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A.D. Chandankhede, K. M. Khandarkar, T. N. Sheikh, R. Joshi and R. S. Dongre

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A Review

Advances in Green Synthesis of Mixed-Metal Metal-Organic Frameworks for Wastewater Remediation

A. D. Chandankhede[1†], K. M. Khandarkar[2], T. N. Sheikh[3], R. Joshi[4] and R. S. Dongre[5]

[1]PGT Department of Chemistry, RTM Nagpur University, Nagpur, Maharashtra, India; dc.aisha1999@gmail.com

[2]Department of Engineering Sciences & Humanities, St. Vincent Pallotti College of Engineering & Technology, Nagpur, Maharashtra, India; kirankhandarkar@gmail.com

[3]PGT Department of Chemistry, RTM Nagpur University, Nagpur, Maharashtra, India; sheikhtahir878@gmail.com

[4]Humanities Department, VMIT Engineering College, Dongergaon, Nagpur, Maharashtra, India; rashmi.joshi13@yahoo.in

[5]PGT Department of Chemistry, RTM Nagpur University, Nagpur, Maharashtra, India; rsdongre@hotmail.com

†Corresponding author: Aisha D. Chandankhede; PGT Department of Chemistry, RTM Nagpur University; dc.aisha1999@gmail.com

Abstract:

Water pollution due to various pollutants is a serious global problem for the environment. Heavy metals, dyes and pharmaceuticals are the main pollutants of water. Traditional techniques are insufficient in removing these complex and persistent pollutants, necessitating the development of advanced materials for the efficient decontamination of water. Metal-

organic frameworks are considered as advanced materials owed to their superior structural and physiochemical properties compared to other conventional materials. Mixed-metal MOFs have shown enhanced properties than monometallic MOFs due to synergistic effect of metal centers. However, conventional MOFs synthesis techniques often involve elevated temperature and pressure, high energy requirement and hazardous solvents which can reduce the environmental sustainability. Green synthesis can provide promising approaches to synthesize mixed-metal MOFs (MM-MOFs) with enhanced physicochemical properties from the synergistic effect of metals. This review provides recent advances in green synthesis of mixed-metal MOFs including microwave-assisted, ultrasonication, mechanochemical and aqueous and room-temperature synthesis. The structural features and advantages of mixed-metal MOFs along with their application in wastewater remediation, particularly for the removal of dyes, heavy metals and pharmaceuticals are discussed. Finally, current challenges are discussed along with future research directions toward sustainable and practical wastewater treatment.

Keywords: Metal-organic frameworks; Mixed-metal MOFs; Green synthesis; Wastewater remediation; Adsorption

1. INTRODUCTION:

Water pollution have become a very serious problem for the environment and living organisms due to growth in population, urbanization and modernization. When the wastewater and the effluent from the industrial, agricultural, and domestic activities are poorly disposed into water bodies, contamination of water takes place which leads to pollution of water. The polluted water contains a wide range of pollutants like heavy metals, inorganic compounds and organic pollutants which can cause adverse effects on human health like neurological disorders, cancer, reproductive problems and short and long-term waterborne diseases. Therefore, wastewater

treatment is necessary for sustainable environment and public health.(Afolalu et al., 2022; Raghavendra, 2024; Saini et al., 2024).

Conventional wastewater treatment includes techniques like aerobic, anaerobic, biofilm technologies, membrane process technologies and advanced oxidation processes which suffer from many limitations like requirement of large land, high capital and operating costs, high energy consumption, membrane clogging, additional requirement of treatment, need skilled supervision and limited effectiveness in handling complex contaminants. Due to these problems there is the need for advanced and sustainable materials for wastewater treatment (Sangamnere et al., 2023).

MOFs are a class of porous materials with large surface area that enhances the adsorption and provides active sites for the removal of contaminant. Furthermore, their pore sizes are tunable due to which the selective pollutant can be removed. By using different combination of metals and organic linkers we can design MOFs with the desired application. The functional groups (NH2) in the framework can strengthens the interaction with pollutants. These advantages have made MOFs as advanced material for efficient remediation of wastewater (Motshekga et al., 2024). The incorporation of two or more metal centres into a MOF can modify the nature and distribution of active sites and alter the local coordination environment. Interactions between the incorporated metals may facilitate adsorption and catalytic processes, resulting in improved performance in suitably designed mixed-metal systems. However, these effects depend on the metal combination, composition, and framework structure. The properties of these MOFs like pore size and electronic properties can be precisely adjusted for a wide range of applications. Consequently mixed metal MOFs are the next generation materials for efficient wastewater remediation applications (Kayani, 2024).

Mixed metal MOFs are very promising in wastewater remediation, however, their conventional synthesis methods have challenges. The traditional strategies need expensive precursors, high temperature and pressure and large amount of organic solvents, which leads to high energy consumption and potential environmental concerns. Furthermore, There are challenges in scalability and maintaining the quality because MOFs are highly sensitive to synthetic conditions making it difficult to reproduce in batch to batch for industrial applications. These limitations can be address by green synthesis which are lower-energy methods and environmentally benign solvents are used (Kumar et al., 2020; Xue et al., 2026). Green synthesized mixed-metal MOFs will combine the advantage of eco-friendly fabrication with enhanced adsorption and catalytic performance (Fig1.). This review focusses on recent advances in green synthesis of mixed-metal MOFs and their applications in wastewater remediation. Furthermore, the challenges in stability, scalability and their applicability in practical world are discussed along with the future directions in this research field. This review will facilitate in developing mixed metal MOFs for wastewater remediation.

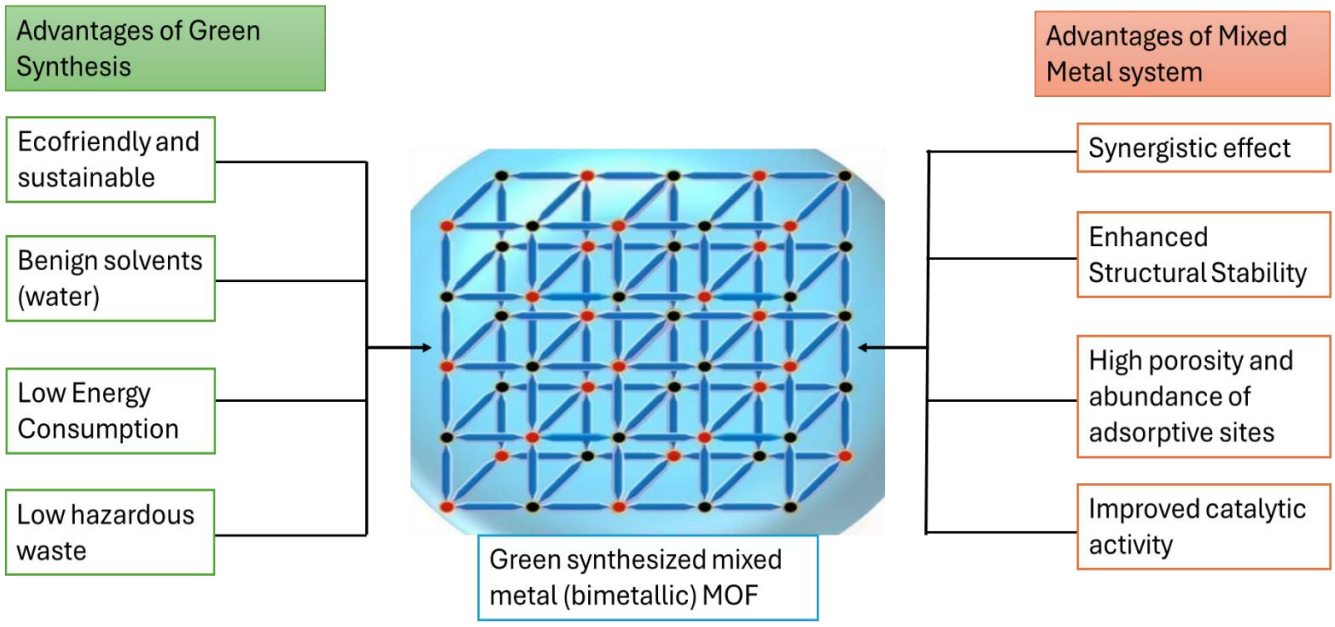


Fig 1. Schematic representation of the combined advantages of green synthesis and mixed-metal systems in Green synthesized Mixed Metal MOFs.

2. METHODOLOGY:

This review examined selected studies on greener synthesis methods and wastewater-remediation applications of mixed-metal MOFs. Relevant literature published mainly during the last ten years, with particular emphasis on studies from the past five years was considered. Relevant literature was identified using combinations of keywords such as “mixed-metal MOFs”, “bimetallic MOFs”, “green synthesis”, “sonochemical synthesis”, “microwave-assisted synthesis”, “mechanochemical synthesis”, “wastewater treatment”, “dye removal”, “heavy metal removal” and “pharmaceutical removal”. Approximately 60 publications were checked for their relevance to this review and around 50 are included in the final manuscript. Studies were included when they reported mixed-metal MOFs, described synthesis approaches relevant to greener preparation such as aqueous, room-temperature, microwave-assisted, sonochemical or mechanochemical routes and investigated removal or degradation of pollutants from water. Relevant review articles were also considered where they helped provide background and current research challenges. Studies were excluded when they dealt with monometallic MOFs or focused on applications unrelated to wastewater treatment. The selected studies were critically examined with respect to metal composition, synthesis conditions, green aspects of the preparation method, structural and physicochemical characteristics, pollutant type, removal mechanism, and remediation performance. The extracted information was subsequently organized thematically to compare synthesis strategies, material properties, and wastewater-remediation applications and to identify current challenges and future research opportunities.

3. FUNDAMENTALS OF METAL ORGANIC FRAMEWORKS:

Metal organic frameworks are porous materials belonging to coordination polymers that are made up of metal nodes and organic linker. MOFs are well defined crystalline structures with

tunable pores and exceptionally high surface area which makes them useful in many applications like adsorption, energy storage, catalysis, and environmental remediation. Metal nodes and organic ligands are the building units of MOFs that are connected through coordination bonds. Metal nodes can be metal ions like Zn^{2+}, Cu^{2+} or polynuclear clusters like Mn_8O_{38} cluster while organic linkers are multidentate ligands which may have functional groups like carboxylate or pyridyl groups. This building units assemble into one-, two-, or three-dimensional frameworks which are highly ordered due to repetition of arrangement throughout the crystal. The physicochemical properties can be regulated by selecting different metal nodes and functionalized ligands. The topology of the framework, pore size and shape can be systematically modify by adjusting synthetic parameters like temperature, solvent identity, pH, and reaction time (Muratov et al., 2024; Terrón et al., 2025; Xue et al., 2026). One of the most distinctive characteristics of MOFs is their exceptionally large surface area which results from the coexistence of well-ordered micropores and mesopores that facilitates mass transport. Due to these features, MOFs have excellent adsorption capacities and catalytic performance for pollutant degradation.(Xue et al., 2026)

The strong tunability of the MOF structure has further led to the development of mixed-metal MOFs, in which multiple metals are incorporated within a single framework (Fig 2). The incorporation of multiple metals can modify the electronic structure, redox behaviour, charge transfer depending on the composition of the framework, ratio and spatial distribution of the metals. Mixed-metal MOFs have demonstrated improved adsorption or catalytic performance toward various contaminants compared with their corresponding monometallic analogues due to increase in the number and diversity of accessible active sites and enhanced oxidation reactions. Synergistic interactions between the different metal can improve the stability of MOFs to withstand the extreme conditions which can improve the practical applicability of mixed-metal MOFs where the real wastewater contains variable pH, ionic strength and other

chemically demanding conditions (L. Chen et al., 2020; Kayani, 2024, 2025; Kayani & Mohammed, 2025; Liu et al., 2022a).

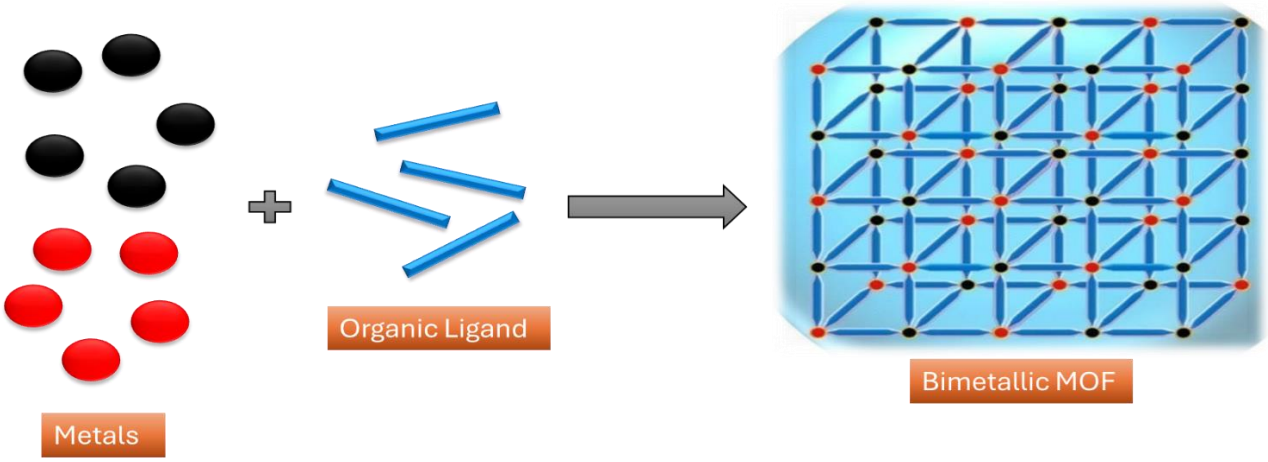


Fig 2. Formation of bimetallic MOFs from two different metals and organic ligand

4. GREEN SYNTHESIS STRATEGIES FOR MIXED-METAL MOFS:

Conventional synthesis of mixed-metal MOFs often involves organic solvents, elevated temperatures and prolonged reaction times. To reduce these limitations, alternative approaches such as ultrasonic, microwave-assisted, mechanochemical, room-temperature and aqueous synthesis have been explored. The sustainability of these methods depends on the overall preparation process, including the solvent system, reaction conditions, washing, activation, and post-synthesis treatment.

4.1. Microwave-Assisted Synthesis

In Microwave-assisted techniques MOFs are synthesized under irradiation of microwaves which directly interacts with the solvent and species in it and causes heating in the medium. This method provides very rapid heating, uniform nucleation and enhances crystallinity, stability, porosity, and functional properties of MOFs. Microwave synthesis is faster than

conventional heating but slower than ultrasound and produces smaller, more homogeneous crystals (Phan et al., 2023).

Several mixed-metal MOFs have been synthesized by microwave assisted methods. For example, Zn/Co-based ZIFs with varying Zn/Co molar ratios were synthesized by microwave-assisted method and applied in visible-light photocatalytic degradation of Congo red. Zn/Co ZIFs were prepared in a very short time with high specific surface areas reaching 1637 $m^2\ g^{-1}$. The incorporation of Zn into the Co-based framework, ZIF-67 was found to enhance the photocatalytic activity in the visible region and the Zn/Co molar ratio of 2/8 exhibited the excellent performance in photocatalytic degradation of Congo red (Tu et al., 2019). Zeng et al. prepared bimetallic Ce-Al-MOFs by microwave-assisted synthesis for phosphate removal from aquaculture effluent. Different molar ratios of Ce:Al:BDC and reaction times were investigated and among them Ce_3Al_3-BDC_3 showed the best phosphate adsorption capacity of 136.99 mg P g^{-1} and maintained effective removal over a broad pH range. The MOFs showed good selectivity for phosphorus even in the presence of co-existing anions and also retained more than 65% phosphate removal efficiency after six adsorption–desorption cycles. Field tests in coastal areas and indoor aquaculture water tests achieved phosphate removal efficiencies above 97%, demonstrating effective performance in real aquaculture water environments. However, the synthesis involved DMF, microwave irradiation at 100 °C and 240 W, solvent washing, and subsequently vacuum-dried at 80 °C. Although microwave heating reduced the reaction time, the use of DMF together with solvent-intensive washing and drying steps limits the overall sustainability of the process.

4.2. Ultrasonic Synthesis:

Ultrasonic synthesis uses the power of ultrasonic waves to synthesize MOFs. It is also referred as sonochemical method where high-frequency sound waves are utilized to generate localized

high-energy conditions in the medium through acoustic cavitation that facilitate rapid and efficient chemical reactions which can significantly reduced the reaction time (Han et al., 2025). The crystal size of MOFs decreases up to nanometer in both sonication and microwave-assisted synthesis that could be beneficial for various applications related to nano-materials (Khan & Jhung, 2015).

Two-dimensional Zn/Co ZIF-L were synthesized in 15 minutes via ultrasonication, whereas the corresponding room-temperature synthesis required about 3 hours. Further TiO_2 nanoparticles are incorporated into Zn/Co ZIF-L via ultrasonication and evaluated the resulting TiO_2@Zn/Co ZIF-L for methyl orange degradation under solar irradiation. After 90 min, Zn/Co ZIF-L and TiO_2@Zn/Co ZIF-L achieved approximately 59% and 84% degradation, respectively. This study indicates that ultrasonication is an efficient method to synthesize mixed-metal MOFs and can introduce nanoparticles within the MOF structure in a short time (Gholampour & Eslamian, 2020). Yao et al. (2021) developed a rapid ultrasound-assisted method to prepare Zn/Co-based zeolitic imidazolate framework nanoparticles. ZIF-8, ZIF-67 and cobalt doped ZIF-8 were synthesized using an ultrasonic bath for 16 min at room temperature. In this study a series of cobalt-doped ZIF-8 materials with different Co ratios was synthesized and investigated the influence of cobalt ion doping on particle sizes which were ranging from 35 nm to over 300 nm due to differences in nucleation and growth rates of zinc and cobalt ZIFs. When applied in heterogenous catalysis for the activation of peroxymonosulfate (PMS) in Rhodamine B degradation, bimetallic Zn/Co-ZIF showed higher catalytic performance than ZIF-8 as well as improved structural stability compared with monometallic ZIF-67. However, the synthesis used methanol in the reaction, the solvent use should also be considered alongside synthesis time and temperature when evaluating the sustainability of the method (Yao et al., 2021).

4.3. Mechanochemical Synthesis:

Mechanochemical synthesis involves the use of mechanical energy to induce the reaction of metals and organic ligands with little or no solvent through ball milling or grinding in mortar and pestle. Due to reduction of solvent, this technique can minimized the negative effects of environment (Han et al., 2025; Głowniak et al., 2021; D. Chen et al., 2019).

Ayoub et al. (2019) reported a target-oriented mechanochemical route for the preparation of a series of mixed-metal MOF-74 materials with controlled metal composition. The approach involved the formation of metal–linker precursors followed by reaction with a second metal source through liquid-assisted grinding, using only a small quantity of methanol during milling. Twelve bimetallic MOF-74 systems were obtained with compositions close to the targeted 1:1 metal ratio, and phase formation was confirmed by PXRD. After activation, the materials retained high porosity, with BET surface areas in the range of approximately 770–1315 $m^2 g^{-1}$. Although the mechanochemical route reduces the amount of solvent required during framework formation compared with conventional solution-based synthesis, the overall process is not entirely energy efficient. During the activation of the materials prior to characterization, the samples were subjected to Soxhlet extraction with hot methanol for 24 h to remove residual linker, followed by outgassing and gradual heating to 150 °C for 5 h. These post-synthesis steps add both solvent and energy requirements to the preparation process. Thus, the main environmental advantage of the method lies in reduced solvent use during the synthesis stage, while the complete process still requires substantial optimization of washing and activation conditions to improve its overall sustainability (Ayoub et al., 2019).

A solvent-free mechanochemical route was also reported for bimetallic CoZn-ZIFs using only powder raw material ZnO, cobalt acetate, and 2-methylimidazole. The Co/Zn composition could be adjusted by varying the amount of cobalt acetate and showed enhanced CO_2 uptake

compared with the Zn-ZIF. This study provides useful evidence that mixed-metal MOFs with tunable composition can be produced through a solvent-free mechanochemical route (Imawaka et al., 2019).

4.4. Room Temperature and Aqueous Synthesis:

Conventional MOF synthesis methods commonly involve toxic organic solvents like dimethylformamide (DMF), elevated temperatures and prolonged reaction times that results in high energy consumption, solvent waste and potential environmental and health concerns. Due to these problems, the recent research on MOF synthesis have shift towards green synthesis strategies that are based on the principles of green chemistry with emphasis on reducing energy requirements and toxic solvents. Aqueous-phase synthesis employs water as the sole or predominant solvent that replaces the toxic organic solvents, while the room-temperature synthesis minimizes the energy consumption in the MOF synthesis (Manikandan et al., 2025; Xue et al., 2026).

Several recent studies have demonstrated the preparation of mixed-metal MOFs at room temperature for removing pollutants from water. For example, ZnNi-MOF-74 and ZnMg-MOF-74 were developed via room temperature synthesis and investigated as adsorbents for Pb^{2+} remediation. These mixed-metal MOF-74 materials have demonstrated the possibility of obtaining functional adsorbents without the prolonged high-temperature conditions associated with conventional MOF synthesis (Flores et al., 2026). Vatani et al. developed Zn/Co bimetallic ZIFs with different metal ratios at room temperature. The metal salts were dissolved in deionized water, while 2-methylimidazole was introduced in an aqueous ammonium hydroxide solution, and framework formation occurred after approximately 10 min of stirring. Variation of the Zn:Co ratio affected the surface area, porosity and optical properties of the resulting

materials. Among the investigated compositions, the Zn:Co ratio of 25:75 showed the best overall water-treatment performance, reaching approximately 97% Cu2+ removal and also exhibiting photocatalytic activity toward methylene blue. These results illustrate the possibility of preparing compositionally tunable bimetallic ZIFs under mild and predominantly aqueous reaction conditions. However, the product was subsequently washed with methanol, kept in methanol for one day and dried at 60 °C for 12 h (Vatani et al., 2024). In another study, Flores et al. (2024) synthesized ZnCu-MOF-74 at room temperature for 20 hours and evaluated it as an adsorbent for removing tetracycline. The room-temperature reaction condition avoids the high energy requirement that is associated with the synthesis involving elevated temperature but used DMF as the solvent. The isolated product was additionally washed with DMF and methanol and soaked in methanol for six days with periodic solvent replacement (Flores et al., 2024). Overall, room-temperature and aqueous synthesis can reduce the thermal and solvent burden associated with conventional MOF preparation but their environmental advantage varies with solvent use, processing time, and subsequent washing and activation requirements.

5. APPLICATION OF MIXED-METAL MOFS IN WASTEWATER REMEDIATION:

The discharge of industrial, agricultural, and domestic effluents containing various pollutants like organic dyes, heavy metal ions, pharmaceuticals, pesticides, and other emerging pollutants in the water bodies has resulted water contamination due to accumulation of these pollutants. The contaminated water can cause threat to human health and ecosystem because of their toxic nature and bioaccumulation. Conventional wastewater treatment technologies are insufficient in cleaning water due to limitations like low selectivity, incomplete pollutant removal, high energy consumption, and the generation of secondary waste. Therefore researchers are focusing

on development of efficient and sustainable materials for water purification. Mixed-metal MOFs have attracted considerable attention for wastewater remediation because of their high surface area, tunable pore structure, and the presence of abundant active sites for pollutant adsorption and catalytic degradation. The incorporation of two or more metal centres within a single framework can alter the local coordination environment and increase the diversity of active sites, which may enhance adsorption, catalytic activity, and structural stability through synergistic interactions between the metals. However, these effects depend on the metals and their composition ratio, coordination environment and synthesis method (Kayani, 2024, 2025; Kayani & Mohammed, 2025; Liu et al., 2022b; Yadav et al., 2026).

5.1. Dye Removal:

Dyes are the most common pollutants that are found in the effluents of the industries like textile, paper, leather, printing, and food industries. They have very complex aromatic structures, high chemical stability and due to this they are very low biodegradable. Thus conventional treatment methods are less effective in removing these complex pollutants which can lead to long-term environmental and health concerns. Mixed-metal MOFs have shown considerable potential for removing both cationic and anionic dyes from water through adsorption and photocatalytic degradation. The dye removal depends on the interaction between the pollutant molecules and the framework, including pore accessibility, surface charge, and the nature of the metal sites (Beydaghdari et al., 2022; Kayani, 2024; Sağlam et al., 2023; Sriram et al., 2022)

The main mechanism in the dye removal is adsorption. Depending on the interaction between the adsorbent i.e. mixed-metal MOFs and the adsorbed dye molecules, the adsorption can occur in two ways: physisorption and chemisorption. Physisorption is the reversible process which involves electrostatic interactions, hydrogen bonding, π-π stacking and van der Waals forces.

While chemisorption is irreversible where strong covalent bonds are formed between the adsorbent and adsorbate. Another promising method is the photocatalytic degradation for the breakdown of complex dye molecules into less harmful compounds. Mixed-metal MOFs can generate reactive species like hydroxyl radicals and superoxide anions under the light irradiation which oxidize and decompose the dye molecules (Kayani, 2024; Sriram et al., 2022; Uddin et al., 2021; Wang et al., 2020).

A green-synthesized bimetallic ZnNi/ZIF-8 nanocrystals exhibited a maximum adsorption capacity of 458 mg g^{-1} toward Acid Blue 92 with a removal efficiency of 93.6%, significantly outperforming pristine ZIF-8. The improved performance was attributed to the synergistic effect of Zn and Ni, which increased surface charge density and promoted multiple adsorption interactions, including electrostatic attraction, hydrogen bonding, hydrophobic interactions, π–π donor–acceptor interactions and van der Waals forces (Soleimani et al., 2025). A carboxymethyl cellulose-supported MIL100(Fe)/MIL88A(Al) composite was synthesized without using toxic solvents demonstrated 738.5 mg g^{-1} adsorption capacity for Reactive Red 198(RR198) along with 87 % photocatalytic degradation of a 100 mg/L tetracycline (TCN) solution under visible light irradiation within 120 min. The synergistic effects of the two MOFs (MIL100(Fe) and MIL88A(Al)) along with CMC enhances active site density with superior structural and chemical stability in aqueous environments and the photocatalytic activity is also enhanced due to formation of a Z-scheme heterojunction that promotes effective separation of photogenerated electron-hole pairs (Rabeie & Mahmoodi, 2025). Mixed-metal MOFs can also act as photocatalyst in visible light for dye removal. The MOF-on-MOF, ZIF-8-on-MIL-10 (ZM composites) were investigated as environmentally friendly photocatalysts for the degradation of Methylene Blue under visible light. Among the synthesized ZM composites, ZM11 had the highest MB photocatalytic degradation of 86.3% and 76% dye removal from water after 3 cycles of MB degradation (Hosseinian Naeini et al., 2024). These studies show

that combining different metal or MOF components can enhance dye removal performance, although the extent of improvement depends on the composition and structure of the material.

5.2. Heavy Metals Removal:

Various industrial processes like mining, electroplating, battery manufacturing, metallurgy, tanneries and fertilizer production releases the effluents that contains heavy metals such as Pb^{2+}, Hg^{2+}, Cd^{2+}, Cr (VI), Cu^{2+}, and As (V) are very toxic and harmful. Unlike many organic pollutants, these toxic heavy metals are non-biodegradable, persistent, and readily bioaccumulate in the food chain that disrupts ecological balance and reduces water quality. Exposure to these toxins can cause neurological, renal, hepatic, cardiovascular, and carcinogenic effects which can be harmful for the human health and aquatic ecosystems. Adsorption is recognized as the most effective technique for removing heavy metal from water because of its simplicity, design flexibility, and can produce high-quality treated water. Many adsorbents can be regenerated through cost-effective and efficient desorption processes which allows its repeated use (Aziz, Mustafa, Hamarawf, et al., 2025; Kayani & Mohammed, 2025; Kobielska et al., 2018; Lin et al., 2023).

Several green-synthesized mixed-metal MOFs have demonstrated excellent removal of toxic heavy metals from the wastewater. A Fe/Co bimetallic MOF nanocatalysts was found to remove heavy metals Pb^{2+}, Hg^{2+}, Cd^{2+}, and Cu^{2+} with removal efficiencies of 98.89%, 95.98%, 93.89%, and 92.60%, respectively, at optimal pH values (5 for Pb^{2+}, 6 for Cu^{2+}/Hg^{2+}, 7 for Cd^{2+}). The bimetallic framework reached adsorption equilibrium within 120 min which is much faster than the corresponding monometallic Fe-MOF and Co-MOF i.e. 6 hours. This indicates that the synergistic interaction between Fe and Co accelerated metal-ion uptake while maintaining structural stability after adsorption (Alshorifi et al., 2022). The beneficial effect of incorporating a second metal is further demonstrated by room temperature synthesized ZnNi-

MOF-74 and ZnMg-MOF-74 with maximum Pb^{2+} adsorption capacities of 1076 and 892 mg g^{-1} respectively while maintaining excellent stability over a broad pH range of 3–8. The processes followed pseudosecond order kinetics for ZnNi-MOF-74 and pseudofirst order kinetics for ZnMg-MOF-74. Spectroscopic analyses suggested that Pb^{2+} removal occurred primarily through electrostatic attraction and ion-exchange processes, in which Pb^{2+} displaced framework metal ions at open coordination sites (Flores et al., 2026). Although the adsorption mechanisms vary depending on the framework composition and target metal ion, the reported studies indicates that electrostatic attraction between positively charged heavy metal ions and negatively charged functional groups on the MOF surface initiates adsorption, followed by coordination interactions at unsaturated metal sites. Ion exchange also plays an important role, whereby incoming metal ions replace framework metal ions at accessible coordination sites. Van der Waals and Hydrogen bonding interactions may further contribute to adsorption depending on the adsorbate and framework chemistry. The coexistence of multiple metals can create a wider variety of adsorption sites and strengthens these interactions, thereby improving both adsorption capacity and selectivity. In another study, a $Ni_{50}Co_{50}$-LDH/UiO-66-$(COOH)_2$ nanocomposite was synthesized in a water-based system without using hazardous organic solvents in the construction of both MOF and LDH, as well as their nanocomposite. The adsorption process followed Langmuir and pseudo-first-order models with maximum adsorption capacities of 415.3 mg g^{-1} for Cd^{2+} and 301.4 mg g^{-1} for Pb^{2+}. The enhanced adsorption performance was attributed to the combination of layered double hydroxide nanosheets with the mixed-metal MOF, which provided abundant adsorption sites and facilitated rapid metal-ion diffusion (Soltani et al., 2021). These studies demonstrate that mixed-metal MOFs and MOF-based composites can effectively remove heavy metals from water and their adsorption performance depends on the metal composition, adsorption mechanism, and operating conditions.

5.3. Pharmaceuticals Removal:

Pharmaceuticals and personal care products (PPCPs) have emerged as a major class of contaminants of emerging concern due to their continuous release into aquatic environments from domestic sewage, hospital effluents, pharmaceutical manufacturing, livestock farming, aquaculture, and improper disposal of unused medications. Conventional wastewater treatment processes often fail to completely eliminate these biologically active compounds because of their complex chemical structures, high stability, and resistance to biodegradation. Consequently, a wide range of pharmaceutical residues, including antibiotics, analgesics, anti-inflammatory drugs, antidepressants, beta-blockers, and other therapeutic agents, have been frequently detected in surface water, wastewater, groundwater and even in treated drinking water. Their persistence in aquatic environments poses significant ecological and human health risks, including the development of antibiotic-resistant microorganisms, bioaccumulation, endocrine disruption, and toxicity to non-target organisms, even at trace concentrations. Therefore, the development of efficient and sustainable technologies for pharmaceutical remediation has become an important priority in wastewater treatment. Among the various treatment approaches, mixed-metal metal–organic frameworks (MOFs) have attracted increasing attention because the incorporation of multiple metals can enhance the number of active sites, improves structural stability, and promotes both adsorption and catalytic degradation of pharmaceutical pollutants (Abbas et al., 2025; Aziz, Mustafa, & Hama, 2025; Hooriabad Saboor et al., 2022; Kayani, 2025).

Mixed metal MOFs have demonstrated significant potential in removing pharmaceuticals from aquatic solutions through adsorption and photocatalytic degradation mechanisms. Flores et al synthesized ZnCu-MOF-74 at room temperature for removing tetracycline from aqueous

solution. This material exhibited the Langmuir maximum adsorption capacity of 775.66 mg g^{-1} and preserved its adsorption capacity after 4 adsorption–desorption cycles. The adsorption process followed the Langmuir adsorption isotherm and the Elovich kinetic model suggesting a physicochemical process. FTIR and XPS analyses suggested the π–π stacking and metal complexation between the TC molecule and ZnCu-MOF-74 as the primary interactions. The π–π stacking from the π-electron density of the benzene rings and the TC structure was proposed as a dominant interaction. Importantly, ZnCu-MOF-74 showed good chemical stability over a pH range of 4–8 and NO_3^-, SO_4^{2-}, Na^+, Mg^{2+}, Cl^-, and Ca^{2+} ions had negligible influence on tetracycline removal. Overall ZnCu-MOF-74 with outstanding cyclability performance can be applied for efficient and effective removal of TC from water (Flores et al., 2024). In another study ZnCoMOF@PAN, a bimetallic ZnCo-MOF was incorporated onto electrospun polyacrylonitrile nanofibers through a room-temperature in situ growth approach for ciprofloxacin (CIP) removal. The composite exhibited well-defined crystallinity with exceptional BET surface area of 1267.87 m^2/g and maximum adsorption capacity of 872.32 mg/g. The adsorption process was found to be spontaneous and well fitted to the Freundlich model and the pseudo-second-order kinetics model. The high CIP removal was attributed to the synergistic effect of hydrogen bonding, electrostatic attraction, π-π stacking, complexation and hydrophobic interactions. The composite retained more than 90% of its initial removal efficiency for CIP after five consecutive adsorption–desorption cycles and maintained over 90% removal efficiency in various real water matrices, demonstrating great adsorption selectivity and practical application in wastewater treatment (Y. Zhang et al., 2026).

In addition to adsorption, mixed-metal MOFs can also function as heterogeneous catalysts for the removal of pharmaceutical pollutants. Le et al. developed FeZn-ZIFs to remove tetracycline antibiotics (TC) from an aqueous solution by activating peroxymonosulfate (PMS). The removal efficiency of TC at concentration of 50 mg·L^{-1} reached 92% within 30 min at room

temperature. The removal ability of the antibiotic was associated with the advanced oxidant processes in which PMS is activated to generate sulphate free radicals that interacted with the tetracycline antibiotic at the active sites of the catalyst surface. The bimetallic catalyst also exhibited good structural stability and recyclability when the removal efficiency of TC can be maintained at approximately 90% after five cycles. This study also demonstrated the removal of methyl violet 2B dyes (MV) through PDS activation, indicating that the material could be extended beyond pharmaceutical contaminants to other persistent organic pollutants (Le et al., 2023). The synthesis conditions and wastewater-treatment performance of selected mixed-metal MOFs are summarized in Table 1, together with the available information on regeneration, structural stability, and metal leaching.

Table1: Comparative summary of selected mixed-metal MOFs for wastewater remediation

Mixed-metal MOF	**Synthesis Conditions**	**Pollutant**	**Performance**	**Reusability**	**Ref.**
Zn–Zr MOF-1	Hydrothermal, ethanol & DMF, 150 °C, 6 h.	Rhodamine B	97.4% degradation in 60 min	72.0% degradation after 3 cycles, FTIR retained, Metal leaching not reported	(X. Zhang et al., 2022)
Fe/Co Bi-MOF	Solvothermal, DMF, 120 °C, 4 h	Pb^{2+}, Hg^{2+}, Cd^{2+}, Cu^{2+}	98.89, 95.98, 93.89 and 92.60% removal, respectively	Fourth-cycle removal decreased to 60.11, 52.09, 46.89 and 50.31%, respectively; quantitative leaching not reported	(Alshorifi et al., 2022)

FeZn-ZIFs	Room temperature synthesis, methanol, 24 hrs.	Tetracycline, methyl violet	92% TC and 95% MV removal.	About 89% removal after 5 cycles, PXRD and FTIR indicated retention of the framework, quantitative metal-leaching data not reported.	(Le et al., 2023)
ZnCu-MOF-74	Room-temperature synthesis, DMF, 20 hrs. MeOH washing/soaking	Tetracycline	775.66 mg g^{-1} Adsorption Capacity	Capacity retained over 4 cycles; crystalline structure retained; ICP-OES detected 2.06 and 2.37 wt% Cu after adsorption and cycling tests, respectively.	(Flores et al., 2024)
Zn/Co-ZIF (Zn:Co=25:75)	RT aqueous synthesis, H_2O/NH_4OH, 10 min, MeOH used during post-treatment	Cu^{2+}, methylene blue	97% Cu^{2+} removal (10 ppm), Approx. 45% MB removal in 200 min	Reusability and metal leaching not reported	(Vatani et al., 2024)
Co-doped ZIF-8	Ultrasound-assisted; methanol; RT, 16 min	Rhodamine B	Approx. 85% degradation in	Crystallinity retained after 24 h in water;	(Yao et al., 2021)

			90 mins with PMS	quantitative metal leaching not reported	
Ce_3–Al_3-BDC_3 MOF	Microwave-assisted; DMF/water; 100 °C, 3 h	Phosphate	136.99 mg P g^{-1}; >97% removal in aquaculture water	>65% after 6 cycles; Ce and Al concentrations in regeneration eluates were almost below ICP-MS detection limits	(Zeng et al., 2025)

6. CHALLENGES AND FUTURE PROSPECTS:

Despite the promising performance of green synthesized mixed-metal MOFs in removing contaminants from water, there are several challenges that still remain in the wastewater treatment. The major challenges are sustainability and scalability of green synthesis, long-term stability and regeneration, environmental safety and realistic wastewater conditions studies.

Several studies have adopted ecofriendly methods like ultrasonication, microwave-assisted, mechanochemical method, room-temperature and aqueous synthesis to reduce the energy requirement that is needed in conventional high temperature and pressure solvothermal synthesis but the low temperature, short reaction time or an aqueous medium alone does not necessarily describe the overall environmental impact of a synthetic method. Several mixed-metal MOFs are synthesized at room temperature but use DMF as solvent and require long reaction time. This indicates that the temperature reduction alone may not be sufficient to achieve a fully sustainable process (Flores et al., 2024). Despite the rapid formation of MOFs in ultrasonic synthesis, the scalability was not reported in some of the studies. The costs

associated with metal precursors, organic linkers, solvents, purification, activation, regeneration and material recovery should be considered. Future work should evaluate green synthesis more comprehensively by considering solvent use, synthesis time, energy requirement, material yield, washing and activation steps instead of individual one like temperature or solvent. Energy consumption, solvent use, waste generation, reaction yield and reproducibility should be systematically compared among synthesis routes. In order to evaluate that the lab scale synthesized MOFs can retain their environmental and economic advantages at larger scales, scale-up studies along with life-cycle and techno-economic assessments are required.

Although mixed-metal MOFs have shown promising performance for the removal of dyes, heavy metals, pharmaceuticals, and other contaminants but their practical use in wastewater treatment is still at an early stage. Many reported studies evaluate a single pollutant in controlled laboratory conditions but they do not fully represent real wastewater, where multiple pollutants and many competing species may be present simultaneously that can affect removal efficiency. Therefore beyond their high removal efficiency in the laboratory, the practical use of mixed-metal MOFs depends on their performance in real wastewater, selectivity in the presence of competing ions, repeated regeneration, framework stability and metal leaching. The effect of competing ions is an important factor because coexisting species can occupy adsorption sites or alter the interaction between the pollutant and the framework. The effect of coexisting ions and complex water matrices has been examined in only a limited number of studies. ZnCu-MOF-74 maintained its tetracycline adsorption performance in the presence of common ions such as NO_3^-, SO_4^{2-}, Na^+, Mg^{2+}, Cl^-, and Ca^{2+} and remained stable over the pH range 4–8. However, the study was mainly conducted in prepared aqueous solutions and also mentioned direct wastewater treatment for future studies (Flores et al., 2024). The microwave-synthesized Ce–Al-MOF was tested in coastal and indoor aquaculture waters and achieved

more than 97% phosphate removal. The material also showed selectivity in the presence of competing anions, providing stronger evidence of performance under realistic water conditions (Zeng et al., 2025). Future research should place greater emphasis on real wastewater, influence of competing species and mixed-contaminant systems. The MOFs showed the high removal efficiencies initially but maintaining this performance after repeated cycles remains an important challenge. The available studies showed loss of performance during regeneration. The stability of Zn-Zr MOF-1 was maintained after three cycles but it showed decrease in RhB degradation efficiency which was associated with the loss of small amount of catalyst during each regeneration step and blockage of active sites when the pollutants or degradation products gradually accumulate during the repeated cycles on the catalyst surface (X. Zhang et al., 2022). The decrease in adsorption of several heavy metal ions was reported in Fe/Co Bi-MOF after four cycles. The reported deactivation was associated with the presence of nitrates or chlorides from the metal salts solution and the loss of mass during washing (Alshorifi et al., 2022). So, the high initial removal efficiency is insufficient to evaluate the MOFs for their practical application, long-term reuse should also be considered. Future studies should increase the number of regeneration cycles and investigate the causes of performance loss through structural and surface characterization before and after reuse. The regeneration should also be optimized to minimize active-site blockage, framework damage and loss of material during recovery. Along with the removal performance, the environmental safety of mixed metal MOFs should also be consider because it contains multiple metals which can release in prolong water treatment. ZnNi/ZIF-8 have shown structural integrity and minimum Zn or Ni leaching in the repeated cycles but the quantification of metals was not studied (Soleimani et al., 2025). Framework stability and metal leaching should be consider separately because retention of the PXRD pattern or FTIR bands confirms that the main framework structure is preserved, but it does not necessarily demonstrate the absence of metal release into water. For example, ZnCu-

MOF-74 retained its crystalline structure after adsorption and cycling, while ICP-OES analysis still detected Cu in the post-treatment solutions (Flores et al., 2024). Future investigations should combine the metal leaching study with the stability and toxicity assessments during wastewater treatment.

Incorporation of multiple metal ions in mixed-metal MOFs have increased the diversity of possible metal combinations and framework composition. The optimization of metal composition in synthesizing mixed-metal MOFs is very important to consider. The relative amount of the two metals can influence the properties and performance of the resulting material. For example, Zhang et al. (2022) studied the effects of different Zn:Zr molar ratios of Zn-Zr MOFs on the structures and morphologies of the MOFs. It was found that the MOF with Zn:Zr molar ratio of 1:1.5 showed the best photocatalytic activity toward Rhodamine B and also exhibited high specific surface area and large pore volume which provided more reactive sites (X. Zhang et al., 2022). Similarly, among the Fe/Co Bi-MOF nanocatalysts investigated by Alshorifi et al. (2022), 50%Fe:50%Co MOF showed the best catalytic performance (Alshorifi et al., 2022). These studies indicate that the proportion of the metals is also important in the performance of mixed-metal MOFs. Future studies should examine a wider range of metal ratios along with the relationships between metal ratios, surface area, structural characteristics, active sites and removal performance. The integration of computational modelling and artificial intelligence can assist in predicting metal–ligand compatibility and framework properties before experimental synthesis. Such approaches can reduce trial-and-error experiments and support the development of Mixed Metal MOFs with improved stability, selectivity and regeneration performance.

7. CONCLUSION:

Mixed Metal MOFs are versatile materials for wastewater remediation that can be made from different combinations of metals and organic ligand which can modify the structural and functional properties of MOFs. Ecofriendly approaches including ultrasonication, microwave assisted, mechanochemical, aqueous and room temperature synthesis provides promising alternatives to conventional environmental burden MOFs synthesis. However, the environmental advantage of these methods depends on the complete synthesis process, including the solvents used, washing, activation, drying, and post-treatment steps. The literature demonstrates that green synthesized Mixed Metal MOFs have been successfully investigated for removing dyes, heavy metals and pharmaceuticals through adsorption and photocatalytic mechanisms but their practical application in wastewater treatment still requires further evaluation. Most studies have employed single-pollutant model solutions, whereas real wastewater contains competing ions and multiple contaminants. Future research should prioritize evaluation under real-wastewater conditions, assessment of long-term framework stability, regeneration over repeated cycles, and quantitative determination of metal leaching. The challenges related to synthesis sustainability and scalability should also be overcome. Overall, the successful translation of MM-MOFs into sustainable wastewater treatment technologies will require a shift from optimizing removal efficiency alone toward the simultaneous optimization of performance, sustainability, stability, regeneration and practical applicability.

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