Recent Development of Metal−Organic Frameworks for Water Purification

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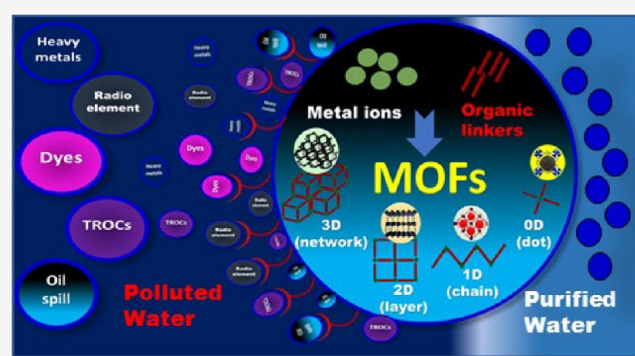Islam M. El-Sewify and Shengqian Ma*

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ABSTRACT: Water contamination is an increasing concern to mankind because of the increasing amount of pollutants in aquatic ecosystems. To purify the polluted water, various techniques have been used to remove hazardous components. Unfortunately, traditional cleanup techniques with a low uptake capacity are unable to achieve water purification. Metal−organic frameworks (MOFs) have recently shown potential in effective water pollutant isolation in terms of selectivity and adsorption capacity over traditional porous materials. The high surface area and versatile functionality of MOFs allow for the development of new adsorbents. The development of MOFs in a range of water treatments in the recent five years will be highlighted in this review, along with assessments of the adsorption performance relevant to the particular task. Moreover, the outlook on future opportunities for water purification using MOFs is also provided.

INTRODUCTION

Water pollution is described as the presence of any foreign components or physical modifications in natural water that either directly or indirectly impact living species (humans, animals, or plants) immediately, after some time, or after a very long period.[1] Water pollution has been caused by an assortment of contaminants, including inorganic, organic, biological, and radioactive toxins,[2] and it is now regarded as one of the most important environmental issues facing the world today. Environmental contamination has been heightened by intensive human activity, such as the use of chemical pesticides and fast industrialization. Various chemical contaminants present in water includes pesticides, halogenated organics, heavy metals, drugs, dyes, and polyaromatic hydrocarbon.[3] Therefore, it has become essential to properly eliminate pollutants from water.[4] In general, there are three basic categories of water purification approaches, such as chemical (oxidation, precipitation), biological (biological aeration filter, activated sludge, etc.), and physical (adsorption technique, membrane filtration).[4] Physical adsorption is a commonly employed technique for achieving extremely low contaminant concentrations. Because of its low cost and excellent adsorption effectiveness, activated carbon is still the most widely used adsorbent in practice today.[5] However, activated carbon's capacity for elimination will significantly decrease over time. Therefore, regeneration after many batches is necessary for recovering its effectiveness in eliminating contaminants.

Because of its low cost and great performance, the adsorption process has been considered to be a promising and feasible approach for effective removal of trace pollutants from various water sources.[6] To enhance the water quality, improved adsorbents with enhanced qualities were engineered to satisfy the water quality requirements. Porous materials with continuous voids in their architectures are thought to be the most suitable basis for designing adsorbents.[7] Many well-known and conventional porous materials, including mesoporous silica,[8] zeolites,[9] and porous carbons,[10] have been investigated for a long time as pollutant adsorbents and have demonstrated sufficient capabilities to separate pollutants from water. Traditional porous materials may exhibit drawbacks, such as limited surface area, less tunable pore structures, and lower adsorption capacity. Compared with traditional porous materials, MOFs have several advantages in the adsorption of water pollutants. MOFs with specific properties have recently

emerged as a novel type of zeolite-like hybrid materials for water purification.[11−14] Typically, they are crystallized compounds that consist of metal ions or clusters and organic, multidentate organic ligands that are held together by coordinate bonds.[15] Some MOFs have remarkably high surface areas and sustained permeability.[16] Since MOFs are often crystalline solids, it is frequently possible to identify and interact with the precise locations of each atom inside the framework with the observed characteristics.[17] MOFs, with their highly customizable pore sizes and greater surface area, offer enhanced efficiency in adsorbing a diverse range of contaminants. Additionally, the tunable functionalities of MOFs enable selective adsorption, which addresses challenges associated with coexisting substances in water. The regeneration and reusability of MOFs stand out in contrast to those of some traditional materials, thereby contributing to sustainable water treatment practices. While cost considerations and scalability challenges may exist, ongoing research aims to overcome these hurdles by emphasizing the unique advantages of MOFs for improved water purification technologies.

The present review aims to provide an overview of the current trend in porous adsorbents as metal−organic frameworks for water purification, as shown in Scheme 1. It will additionally

Scheme 1. General Schematic Representation about the Usage of Different Types of MOFs in Water Purification
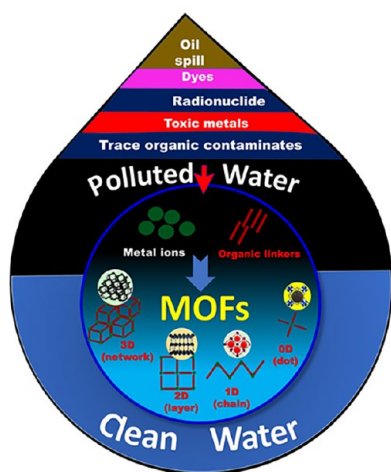
highlight significant developments in the study of MOF-based adsorbents for different types of contaminant sequestration and provide an outlook on the creation of next-generation porous adsorbents. In this review, we intend to give some beneficial suggestions for reducing threats to water quality.

WATER PURIFICATION

Contaminants can be categorized into two subclasses: inorganic and organic, depending on the chemical components of chemicals found in water.[18] For instance, inorganic metal emissions from factories are the main cause of heavy metal pollution, a well-known problem with water quality.[19] These can be extremely hazardous and carcinogenic and endanger the general public's health. Additionally, a number of organic contaminants, mostly from industrial activity and rising plastic product use, have been found in water supplies around the world, including drinking water, in detectable amounts. The rapid buildup of these dangerous substances in human bodies as a result of exposure to contaminated water can have negative

health effects and dramatically raise the chance of contracting various diseases.[20] Although there is still plenty of work to go until large-scale porous adsorbents are actually used in water filtration, metal−organic frameworks show promising possibilities in preventing this unacceptable consequence.[21−23] The stability of MOFs in aqueous environments is a pivotal consideration for their successful application in water purification. Water stability directly influences the durability of MOFs during extended use in water treatment applications. The potential degradation or leaching of the MOF components could compromise their efficiency and pose challenges for large-scale deployment. To comprehensively evaluate the water stability of MOFs, researchers should employ advanced characterization techniques, such as in situ spectroscopy or X-ray diffraction. Moreover, conducting prolonged exposure experiments under conditions mimicking real-world water treatment scenarios is essential. Exploring postsynthetic modifications to enhance the water stability of MOFs is a promising research direction, such as incorporating hydrophobic groups, employing protective coatings, or optimizing metal−linker interactions, can be investigated to mitigate water-induced degradation and improve overall stability. By employing a multidisciplinary approach that combines materials science, analytical chemistry, and environmental engineering, researchers can contribute to the development of robust MOFs capable of withstanding the challenges posed by real-world water treatment.

TREATMENT OF ORGANIC CONTAMINANTS USING MOFS

An emerging promising path in environmental treatment is the use of MOFs for the treatment of organic pollutants. The specific characteristics and high surface area of MOFs are remarkably effective at adsorbing a variety of organic pollutants. Because MOFs are porous, organic molecules can be effectively trapped and removed from water, thereby solving problems caused by contaminants, including dyes, oil spills, pesticides, and pharmaceutical residues. MOFs provide a focused and effective remediation approach because of their ability to be specifically designed to optimize them for particular organic contaminants. MOFs' reversible adsorption mechanism enables regeneration and reuse to be simpler, which supports environmentally friendly and economically viable treatment methods. MOFs' distinctive qualities establish them as viable tools for organic contamination cleanup and provide creative solutions for sustainable development, especially as studies on this sector progress.

Dyes Removal. Artificial dyes have become widely used in modern industry, which has brought down the overall expense of pigmentation technology significantly, as a result of the increasing need for colorful clothing and textiles at affordable prices brought on by the world's population growth.[24,25] There are currently more than 10 000 distinct dyes on the market, and it is estimated that over 700 000 tons of synthetic dyes are consumed annually.[26] A significant percentage of untreated dye effluent is released into surface water, and the quantity of these eliminated dyes amounts to more than 15% of the dyes generated yearly. Dyes are difficult to decompose/degrade after entering water bodies because of their sturdy chemical composition. These persevering dyes will ultimately incorporate into household water, which poses a wide range of major health risks.[27]
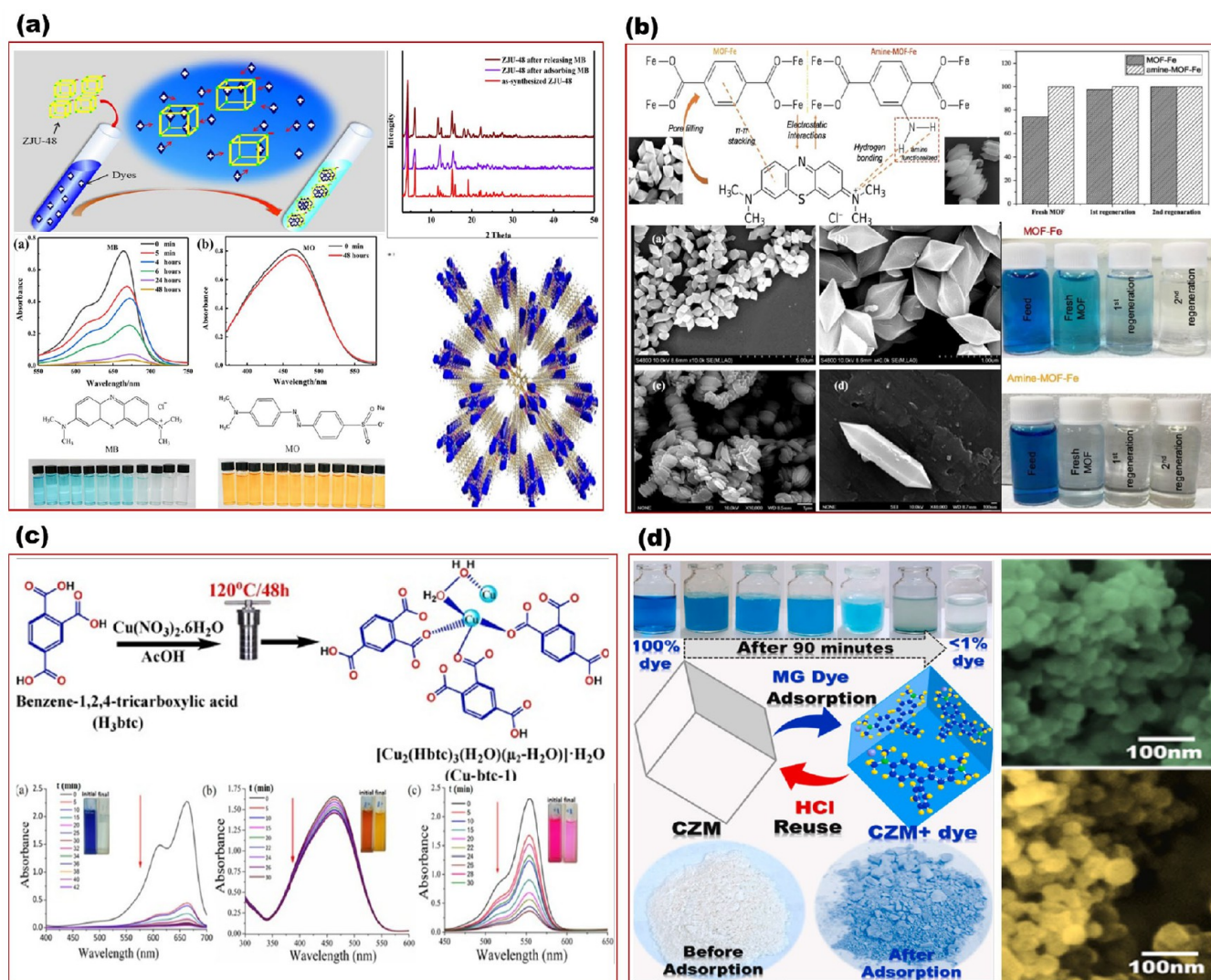
Figure 1. (a) Synthesis of porous MOF ZJU-48 with 1D channels for eliminating cationic dyes rather than anionic dyes.[34] Copyright 2020 American Chemical Society. (b) Preparation of MOF-Fe and amine-MOF-Fe for cationic methylene blue. Reproduced with permission from ref 35. Copyright 2020 Elsevier. (c) Cu-btc-1 was synthesized and characterized for adsorption of dye methylene blue from the mixture of three organic dyes. Reproduced with permission from ref 36. Copyright 2020 Elsevier. (d) Fabrication of cubic Zn metal−organic frameworks (MOFs) for capturing MG dye from wastewater. Reproduced with permission from ref 39. Copyright 2021 Elsevier.

Dye removal using MOFs involves the use of these porous materials to adsorb and remove dye molecules from water or wastewater.[28] The design of MOFs with large surface areas and tunable pore sizes is particularly suitable for the removal of organic dyes. MOFs can be synthesized with specific structures and functional groups to enhance their affinity for certain dyes.[29] When MOFs come into contact with a dye-contaminated solution, they readily adsorb dye molecules within a few minutes. The adsorption mechanism of an MOF for dye removal can be explained with different interactions, including electrostatic forces, hydrogen bonding, and van der Waals forces.[30] Dye molecules can penetrate the pores of an MOF's structure, thereby allowing for higher adsorption capacity, and can be engineered to be selective for specific dyes or classes of dyes (for a summary of dye removal performance of various MOF adsorbents,[31] see Table S1). Colorimetric methods and UV−vis spectroscopy are two approaches that may be employed to determine the amount of dye present in the wastewater. Several parameters, like temperature, pH, and contact duration, may be

used to optimize the effectiveness of eliminating dyes and the adsorption capacity of MOFs.[32] Because of its strong contrast and outstanding visibility, methylene blue (MB), one of the essential cationic dyes, is utilized in the textile industry for materials, including cotton, silk, paper, wool, and wood.[32] Extreme health concerns, such as jaundice, perspiration, heart difficulties, eye irritation, nausea, quadriplegia, sickness, anemia, etc., can result from elevated environmental contact with MB.[33] Therefore, it is crucial to remediate MB-based wastewater. Because of its negatively charged surface, Xie and co-workers developed a porous MOF ZJU-48 with 1D channels that exhibited water stability for eliminating cationic dyes rather than anionic dyes.[34] Furthermore, the adsorptive capacity of the MOF toward dyes reduced as the dye size increased, thereby indicating that ZJU-48 is an excellent adsorbent not only for cationic dyes but also for small organic compounds. Furthermore, the cationic small dye MB had a maximum adsorption capacity of 582.44 mg/g, and approximately 90% of MB could be released from MOF ZJU-48 over 6 h (Figure 1A).
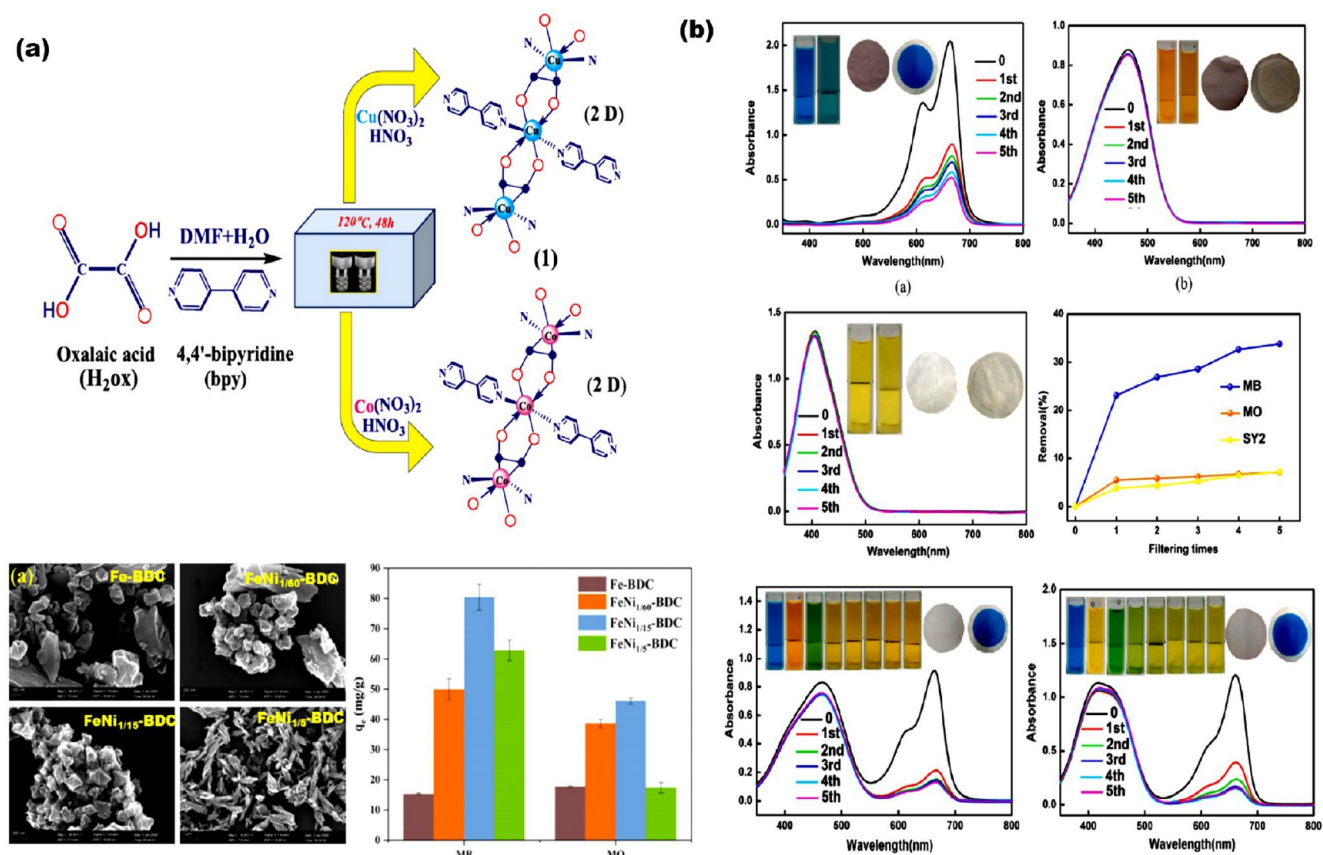
Figure 2. (a) Synthesis of (2D) metal organic frameworks (MOFs Cu-MOFs remove the MB). Reproduced with permission from ref 40. Copyright 2019 Elsevier. (b) Design of ionic bio-MOF-1 using PAN via electrospinning to remove and extract MB. Reproduced with permission from ref 41. Copyright 2019 Elsevier.

The release process of MB dye from ZJU-48 in DMF solution with NaCl was utilized to determine the desorption capacity of the MOF, which showed that ZJU-48 was stable and could be regenerated. Paiman et al. created MOF-Fe and amine-MOF-Fe for cationic dye (MB) elimination with adsorption capacities of 149.25 and 312.5 mg/g, respectively.[35] The effect of amine functionalization demonstrated that the amine group was essential in the adsorption process, which yielded a 2-fold increase in adsorption capacity toward MB ions. The adsorption kinetics of MOF-Fe and amine-MOF-Fe were demonstrated that both MOFs followed the pseudo-second-order kinetics model, and after the release process of MB following two cycles, the amine-MOF-Fe was able to maintain its adsorption capacity (Figure 1b). Mantasha et.al. synthesized and characterized Cu-btc-1, in order to adsorb and separate the dye, methylene blue from the mixture of three organic dyes [methylene blue, methyl orange, and rhodamine B (Rh-B)] in wastewater.[36] The prepared Cu-btc-1 is water stable, recyclable for four times, and selective for removal of MB at optimum conditions with a removal capacity 98.67%. The aromatic moieties of carboxylates of Cu-btc-1 form strong electrostatic interactions, H-bonding, and $\pi-\pi$ interactions with MB$^+$ (Figure 1c). A cationic dye known as malachite green (MG) has a wide range of commercial and industrial uses.[37] MG is utilized in the food sector as a flavoring and coloring agent and in the leather, silk, wool, cotton, paper, and acrylic industries as a dyeing material.[38] In order to remove MG dye from wastewater samples, El-Sewify et al. created cubic inorganic−organic Zn metal−organic framework (MOF) at room temperature using a one-pot synthesis

approach.[39] The nanocubes have homogeneous micropores that could capture MG dye from wastewater and intergrowing chains with highly accessible surface surfaces. The Zn-MOF adsorbent's active, negatively charged surface was produced under optimum conditions. The calculated adsorption capacity of cubic zinc MOF for MG removal was demonstrated by adsorption studies (953.14 mg g^{-1}), and this capacity remained even after several reuse cycles (Figure 1d).

For the purpose of eliminating the organic dyes methyl orange (MO) and methylene blue (MB), Mantasha I. et al. synthesized (2D) MOFs $[M(ox)(bpy)]_n$ $[M = Cu(I)$ or $Co(II)]$ under solvothermal conditions using straightforward and affordable procedures.[40] Adsorption takes place through a pseudo-second-order route, and the electrostatic, $\pi-\pi$, and H-bonding interactions between the MOF and the dye might be a realistic mechanism of adsorption. Under optimal conditions, the Cu-MOFs remove the maximum amount of MB (96−98%). Additionally, even in mixed dye applications, a high selectivity of Cu-MOFs toward MB was reached within 10 min, which signifies the practical applicability of this technology for realistic and long-term use that can be used for five cycles. The adsorption mechanism takes place through electrostatic, $\pi-\pi$, and H-bonding interactions between the Cu-MOF and the dye (Figure 2a). Li et al. successfully fabricated an ionic MOF via electrospinning by selecting polyacrylonitrile (PAN) as the template and encapsulating anionic bio-MOF-1 into the nanofibrous matrix.[41] In contrast, PAN displays a relatively low removal efficiency (33.8%), while Bio-MOF/PAN displays a greater removal percentage (74.6%). The highly specific
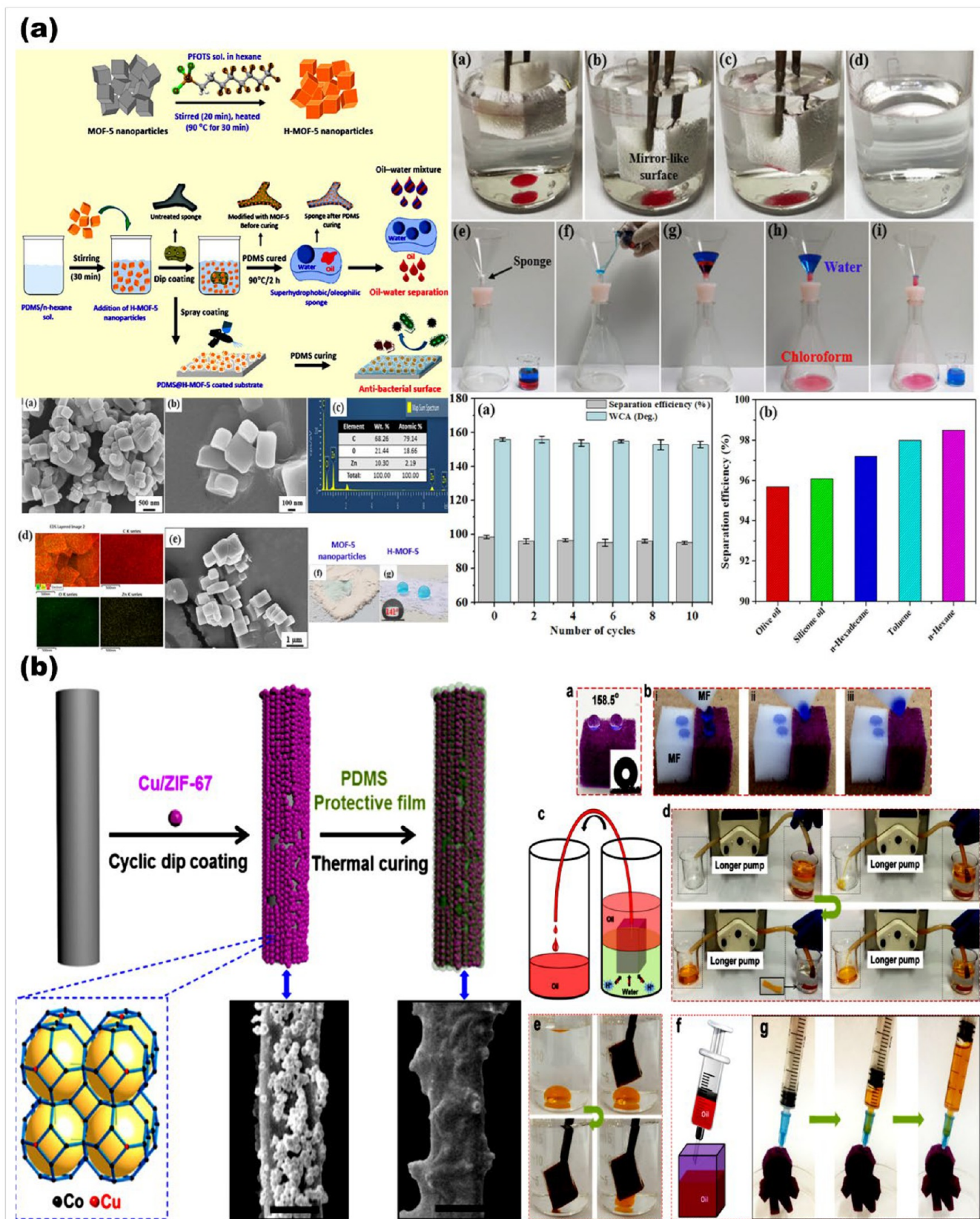
Figure 3. (a) Synthesis of a superhydrophobic/superoleophilic sponge using hydrophobic MOF-5 and then modification of it with PDMS for the separation of oil from water. Reproduced with permission from ref 51. Copyright 2022 Elsevier. (b) Cu/ZIF-67 was fabricated on a PDMS-coated melamine sponge using an immersion technique to create an oil-absorbent material that may be squeezed to discharge the absorbed oil. Reproduced with permission from ref 52. Copyright 2020 Elsevier.
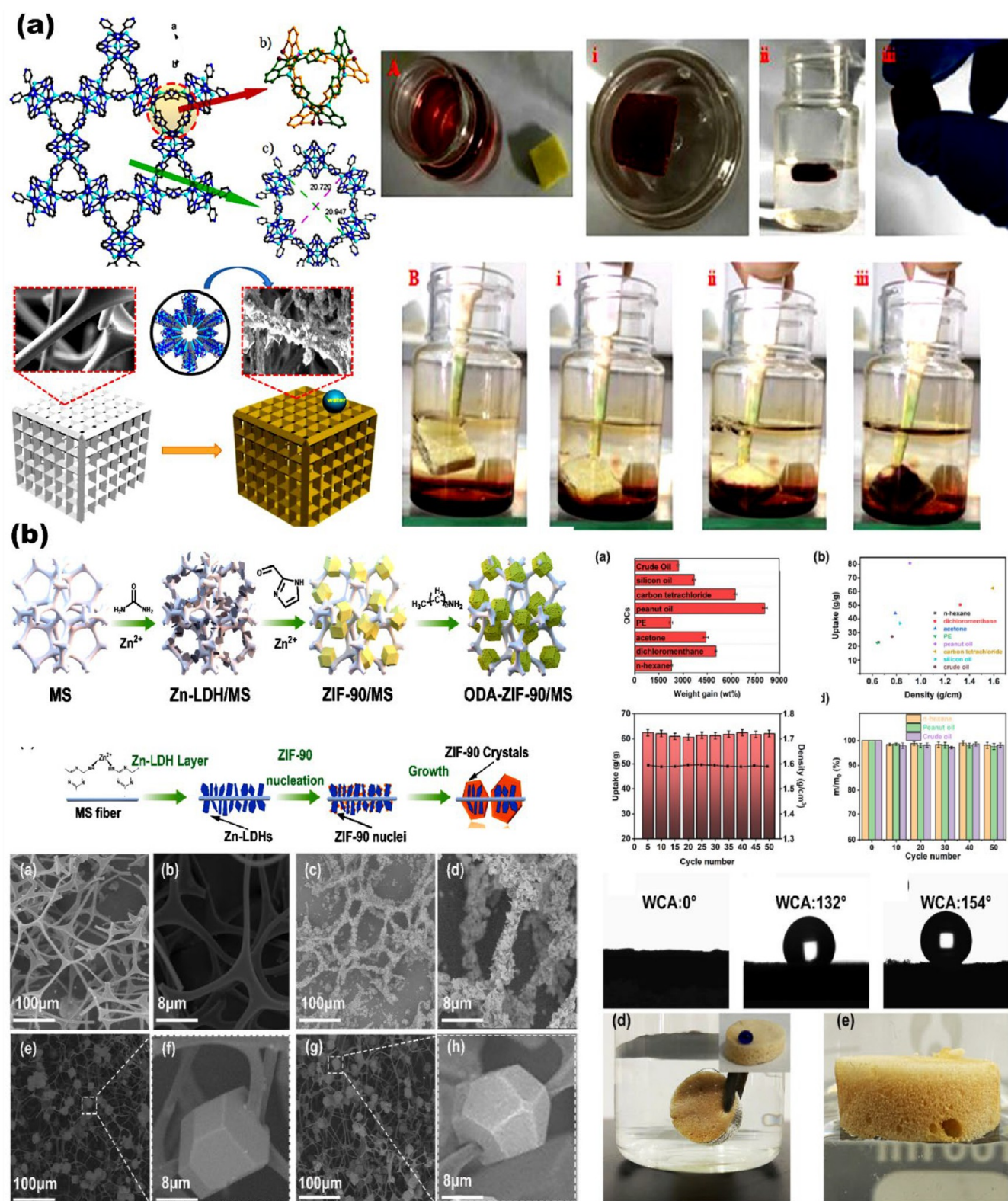
Figure 4. (a) Development of a porous metal−organic framework (HPU-13) with melamine sponge (MS) to remove oil from water. Reproduced with permission from ref 53. Copyright 2019 Elsevier. (b) Fabrication of modified ZIF-90/MS with octadecylamine (ODA) sponge for selective oil−water separation. Reproduced with permission from ref 54. Copyright 2023 Elsevier.

extraction of MB from the combination of several dye solutions is demonstrated by these nanofiber-based MOFs. To prevent bead development during the manufacture of nanofibers, it is a tough goal to optimize the ratio between the polymer solution and the materials. It can make the fiber less effective for the intended use (Figure 2b). After five cycles, the adsorption capabilities of the bio-MOF/PAN filter toward MB+ were generally steady.

Dye removal using MOFs is a promising approach for treating dye-contaminated wastewater, especially in industries, such as textiles, dyes, and pigments. The versatility and tunability of MOFs make them a valuable tool for addressing environmental and water pollution concerns related to dyes.

Oil Spill Cleanup. The world's ecosystems are at risk because of rising concerns about oil leaks from crude oil production and distribution. The annual expense of cleaning up oil spills has been estimated to be above $10 billion worldwide.[42] Oil spill cleaning techniques that rely on antiquated methods, including in situ combustion and centrifugation, are expensive, inefficient, and may cause secondary contamination.[43] MOFs are extremely porous materials that are used in oil spill cleaning procedures to selectively absorb and extract spilled oil from water environments.[44] MOFs with hydrophobic and high surface area characteristics offer several advantages for oil spill cleanup and the ability to be tailored for specific applications.[45] The hydrophobic design of MOFs allows for high affinity for oil molecules in water.[46] The MOF selection with the appropriate pore size, structure, and functional groups is essential for effective oil adsorption. MOFs can be used as powders, pellets, membranes, or coatings on materials like fabric or mesh depending on the nature of the oil spill and the environment.[47] The hydrophobic interactions between the MOF structure and the nonpolar oil molecules cause the MOFs to rapidly capture the oil when they contact the spilled oil. The porous nature of MOFs attracts and traps oil molecules, and MOFs may be engineered to float on the water's surface in a variety of oil spill circumstances. As a result, they can successfully remove oil from the surface of water and restrict its spread. MOFs can be tailored to selectively adsorb certain types of oils or hydrophobic compounds while repelling water[48] (see Table S2). Oil-saturated MOFs are frequently regenerable for further usage via use of vacuum treatment, solvent extraction, or heating. For cleanup efforts to be both sustainable and economical, MOFs must be regenerated. The recovered oil can be separated from the MOFs, and the MOFs can be disposed of or recycled for future use. Proper disposal methods must be followed to minimize the environmental impact. Considerations should be made regarding the overall environmental impact of using MOFs in oil spill cleanup. This includes the energy and resources required for MOF production and regeneration, as well as the disposal or recycling of MOFs. Yang et al. demonstrated the adsorption of common aromatic and aliphatic oil substances using fluorous metal−organic frameworks (FMOFs) with hydrogen-free, fluorine-lined pore surfaces.[49] Because of its hydrophobic pore surface, the synthesized FMOF-1, which was produced out of silver(I) 3,5-bis(trifluoromethyl)-1,2,4 triazolate (AgTz), showed a remarkable ability to clean up oil spills. One of the most common MOFs, such as HKUST-1, was also utilized for oil removal from water.[50] Its ability to remove oil has been assessed in an oil and water solution with 300 mg L^{-1} of oil in the emulsion. Once an emulsion containing the HKUST-1 dispersion was shaken vigorously, a clear solution appeared when the procedure of adsorption was accomplished. HKUST-1 was shown to have a maximum removal capacity of 4000 mg g^{-1}. Great efforts were made to develop MOFs on a range of scaffolds to improve their ability to absorb organic oils. For purification of water from oil-in-water emulsions, a significant number of hydrophobic MOF-based sponges and foams were produced. Using a simple dipping technique, Barthwal et al. constructed a superhydrophobic/superoleophilic sponge by generating hydrophobic MOF-5 and then modifying it with PDMS.[51] The separation efficiency of up to 98% for a variety of oil/water mixtures was demonstrated by the prepared superhydrophobic PDMS@H-MOF-5-coated sponge, which also can be recycled (10 times). The developed PDMS@H-MOF-5 suspension may also be spray-coated onto various surfaces to produce superhydrophobic surfaces (Figure 3a). The Cu/ZIF-67 was successfully fabricated on a PDMS-coated melamine sponge, as demonstrated by Lei et al., using a simple immersion technique.[52] Porous melamine sponge, rough Cu/ZIF-67 particles, and hydrophobic poly(dimethylsiloxane) (PDMS) worked together to create an oil-absorbent material that may be squeezed to discharge the absorbed oil. Because of its low density, the MF-Cu/ZIF-67@PDMS sponge demonstrated an exceptional mass-based oil absorption capacity, with its mass-based absorption capacity ($m_{oil}/m_{absorbent}$) reaching 2000 to 3800 wt % for a variety of organic solvents or oils (Figure 3b). The MF-Cu/ZIF-67@ PDMS sponge maintained 90% of its initial oil absorption capacity after 100 cycles of absorption and squeezing, while the oil residue in this sponge stayed below 10% following squeezing. The MS-CMC-HPU-13 composite was developed by Xu et al. to remove oil from water through the combination of porous MOF (HPU-13) with melamine sponge (MS).[53] The oil can be collected from water with an adsorption capacity of 13 000%. Furthermore, a self-assembly pump made of MS-CMC-HPU-13 was able to remove oil in situ from water consecutively (Figure 4a). Luo et al. fabricated ZIF-90/MS with octadecylamine (ODA) to prepare ODA-ZIF-90/MS composite sponge with enhanced hydrophobic character.[54] The ODA-ZIF-90/MS has a multilevel pore structure and exhibits excellent hydrophobicity, which demonstrated great potential for selective oil−water separation. In addition, it has an absorption capacity of 22−63 times its weight and can be reused with 98.7% absorption retention after 50 cycles (Figure 4b). The use of MOFs in oil spill cleanup provides an innovative and efficient approach to mitigate the environmental impact of oil spills. Oil pollution in aquatic environments may be effectively and sustainably addressed with MOFs because of their ability to regenerate and selectively absorb oil.

Trace Organic Contaminants Treatment. Trace organic contaminants (TrOCs) can include various pollutants, such as pharmaceuticals, personal care products, pesticides, and other organic chemicals that are present in water sources at low concentrations.[55] Freshwater lakes and rivers are frequently contaminated by TrOCs.[56] The primary sources of these contaminants are industrial processes, agricultural production, and wastewater from cities.[57] TrOCs have been identified in the water's surface at low levels of between one part per trillion (ppt) and one part per billion (ppb). However, there has recently been adequate evidence to show that continuous consumption of even extremely minimal quantities of TrOCs will be harmful to human health.[58] Many biological and chemical procedures have been employed over the past few years to separate volatile organic compounds (VOCs) from water.[59] However, as the majority of VOCs are challenging to remove, traditional water purification techniques have not been highly effective. MOFs have excellent potential for the efficient elimination of toxic TrOCs because of their superior adsorption−desorption characteristics.[60] Treatment of trace organic contaminants using MOFs is an emerging area of research and application in environmental remediation.[61,62] MOFs can be engineered for selective adsorption of specific contaminants and it can be achieved through functionalization of MOFs with organic ligands or by selecting MOFs with the appropriate pore size
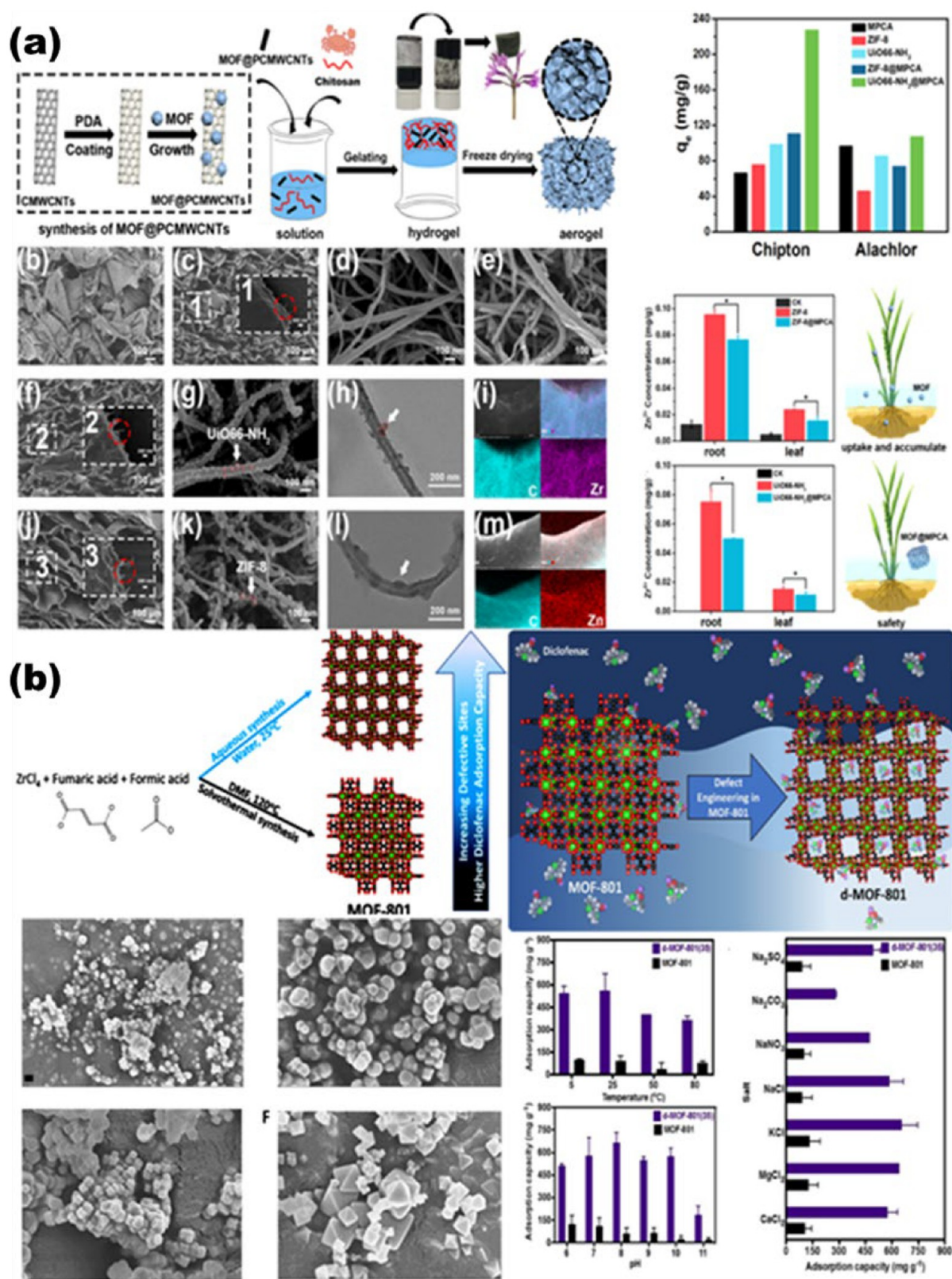
Figure 5. (a) Synthesis of ZIF-8 or UiO66-NH$_2$ onto carbon nanotube aerogels (MPCA) via in situ nucleation and growth of MOFs nanoparticles onto carbon nanotubes for removal of chipton and alachlor. Reproduced with permission from ref 67. Copyright 2021 Elsevier. (b) Synthesis of defective MOF-801 for increased diclofenac adsorption by replacing water with dimethylformamide. Reproduced with permission under Creative Commons CC-BY license from ref 69. Copyright 2022 The Authors.

and chemical properties.[63] The mechanism of trace organic contaminants adsorption onto MOFs is primarily driven by physical interactions, including van der Waals forces, hydrogen bonding, and $\pi-\pi$ interactions.[64] Analytical techniques, such as
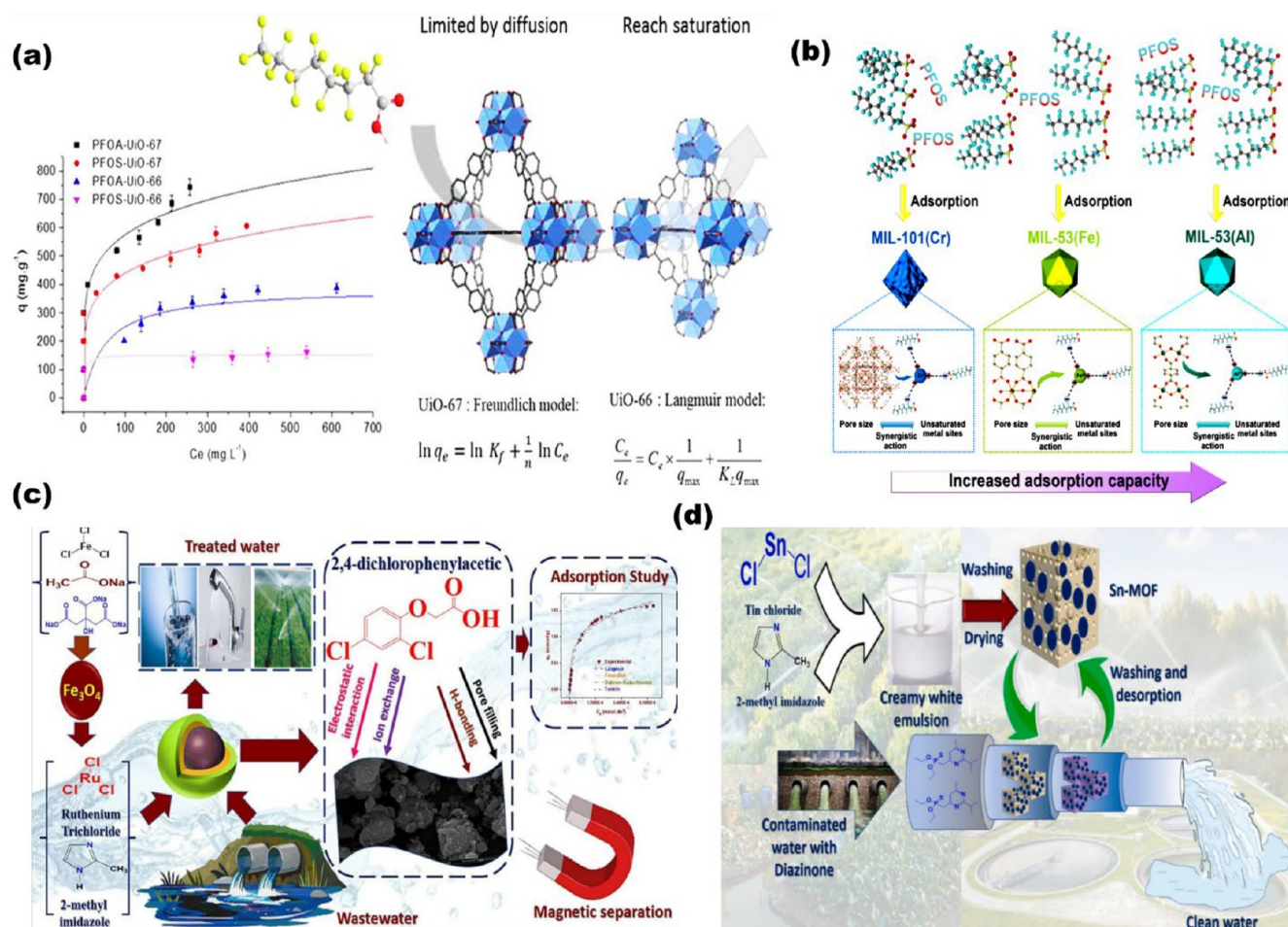
Figure 6. (a) Preparation of zirconium metal−organic frameworks (MOFs, UiO-66 and UiO-67) for different removal processes of PFOS and PFOA. Reproduced with permission from ref 71. Copyright 2019 Elsevier. (b) MIL-53(Al), MIL-53(Fe), and MIL-101(Cr) were utilized for PFOS adsorption from water. Reproduced with permission from ref 72. Copyright 2021 Elsevier. (c) A magnetic ruthenium MOF (MRM) fabricated for eliminating pesticide 2,4-dichlorophenylacetic from aqueous solutions. Reproduced with permission from ref 73. Copyright 2023 Elsevier. (d) Tin−metal organic frameworks (Sn-MOFs) were designed for removing the organophosphate insecticide diazinon (DZ) from water. Reproduced with permission from ref 74. Copyright 2023 Elsevier.

liquid chromatography and mass spectrometry, are used to monitor and confirm the removal of trace organic contaminants from water after MOF treatment. Diffuse chemical contamination that is hard to regulate is largely caused by improper use of pesticides.[65] Therefore, it is critical to remove pesticides from the environment to protect humans and environment. Pesticide usage due to inadequate utilization poses several concerns to the environment, including significant residual chemicals and the deterioration of agricultural environments and soil health.[66] To remove the two regularly used herbicides, such as chipton and alachlor, Liang et al. loaded two commonly employed MOFs (ZIF-8 or UiO66-NH$_2$) onto carbon nanotube aerogels (MPCA) via in situ nucleation and growth of MOFs nanoparticles onto carbon nanotubes.[67] The optimal adsorption capacity of chipton captured by UiO66-NH2@MPCA was 227.3 mg/g and it could be reused five times without significantly reducing the adsorption efficiency. The MPCA demonstrated that the active sites of the exposed surface area had a major impact on the adsorption of chipton or alachlor molecules onto the composite materials. For the chipton, the estimated maximum adsorption capacity (q_m) using the Langmuir model of MPCA, ZIF-8@MPCA, and UiO66-

NH$_2$@MPCA was 158.5, 160.9, and 246.8 mg/g, respectively; for the alachlor, the q_m was 264.6, 196.2, and 232.8 mg/g (Figure 5a). Furthermore, nonsteroidal anti-inflammatory medications (NSAIDs), like diclofenac, are highly prescribed and utilized all over the world.[68] It is crucial to remove these materials from the environment because they pose a threat to aquatic life. Prasetya and co-workers synthesized defective MOF-801 for increased diclofenac adsorption by replacing water with dimethylformamide.[69] The defect concentration in MOF-801 can be rationally tuned by adjusting the modulator concentration (formic acid) in the synthesis solution. The adsorption capacity of highly defective MOF-801 for diclofenac can reach 680 mg g^{-1} and can be regenerated for up to four cycles of the adsorption test (Figure 5b).

The extremely hazardous substances perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA), which are almost not recyclable, cause significant contamination in the environment.[70] Sini et al. employed zirconium MOFs (UiO-66 and UiO-67) to eliminate persistent pesticides (PFOA and PFOS) from water.[71] With a different removal process, UiO-67 exhibits larger adsorption capacities up to 743 mg g^{-1} for PFOA and 585 mg g^{-1} for PFOS compared with UiO-66 (388 and 160 mg g^{-1})
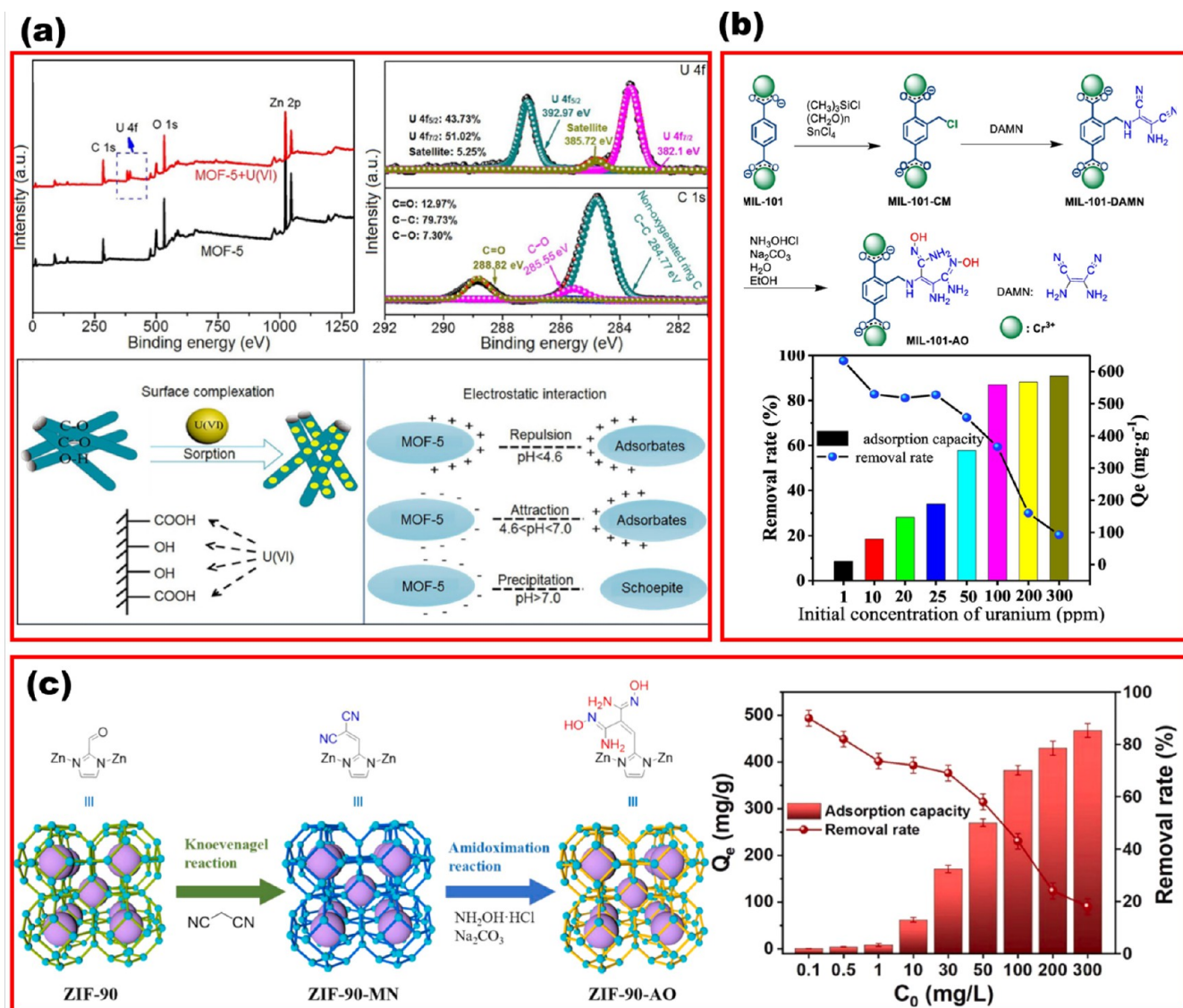
Figure 7. (a) General schematic diagram for manufacturing the rodlike MOF-5, which was then utilized to extract U(VI) from radioactive wastewater. Reproduced with permission from ref 86. Copyright 2018 Elsevier. (b) A postsynthesis approach was employed to add certain active functional groups to MIL-101 to generate modified porous material (MIL-101-AO) for uranium adsorption. Reproduced with permission from ref 87. Copyright 2020 Elsevier. (c) Fabrication of amidoxime-functionalized ZIF-90, ZIF-90-OM, and ZIF-90-AO for the extraction of U(VI). Reproduced with permission from ref 88. Copyright 2022 Elsevier.

for PFOA and PFOS, respectively (Figure 6a). Zhao and co-workers designed MIL-53(Al), MIL-53(Fe), and MIL-101(Cr) for PFOS adsorption from water.[72] Because of its smaller pore size and more unsaturated metal active sites compared with MIL-53(Fe) and MIL-101(Cr), MIl-53(Al) performs better in removal of PFOS. The same group also investigated the possibility of increasing the PFOS adsorption capacity in the presence of Fe^{3+} ions, which results from the contact and bridging of the Fe^{3+} ion with the PFOS molecule (Figure 6b). Al-Ahmed et al. fabricated a magnetic ruthenium metal−organic frameworks (MRM) for eliminating dangerous pesticide 2,4-dichlorophenylacetic (2,4-D) from aqueous solutions.[73] The synthetically created MRM adsorbent has an exceptional yield, ability, and reversibility with up to five cycles of adsorption/desorption. They obtained an adsorption capacity of 2,4-D onto MRM of 293.93 mg g^{-1}, and the MRM could be regenerated for more than five cycles (Figure 6c). The effectiveness of a tin-MOF (Sn-MOF) in eliminating the organophosphate insecti-

cide diazinon (DZ) from water was studied by Alrefaee et al. With an adsorption capacity of 587.39 mg g^{-1}, the Sn-MOF adsorbent demonstrated the ability to adsorb DZ onto Sn-MOF.[74] It can also be cycled up to five times for adsorption−desorption procedures (Figure 6d). MOFs hold promise as a sustainable and effective technology for the removal of trace organic contaminants from water sources.

■ TREATMENT OF INORGANIC POLLUTANTS

The treatment of inorganic contaminants in water using Metal−Organic Frameworks (MOFs) represents a promising frontier in water purification technology. MOFs have a strong attraction toward heavy metals, metal ions, and other inorganic species via processes including ion exchange and coordination. MOFs' flexible characteristics of MOFs enable them to be materials with unique binding sites that are suited to target specific pollutants, thereby providing a flexible and effective method of water
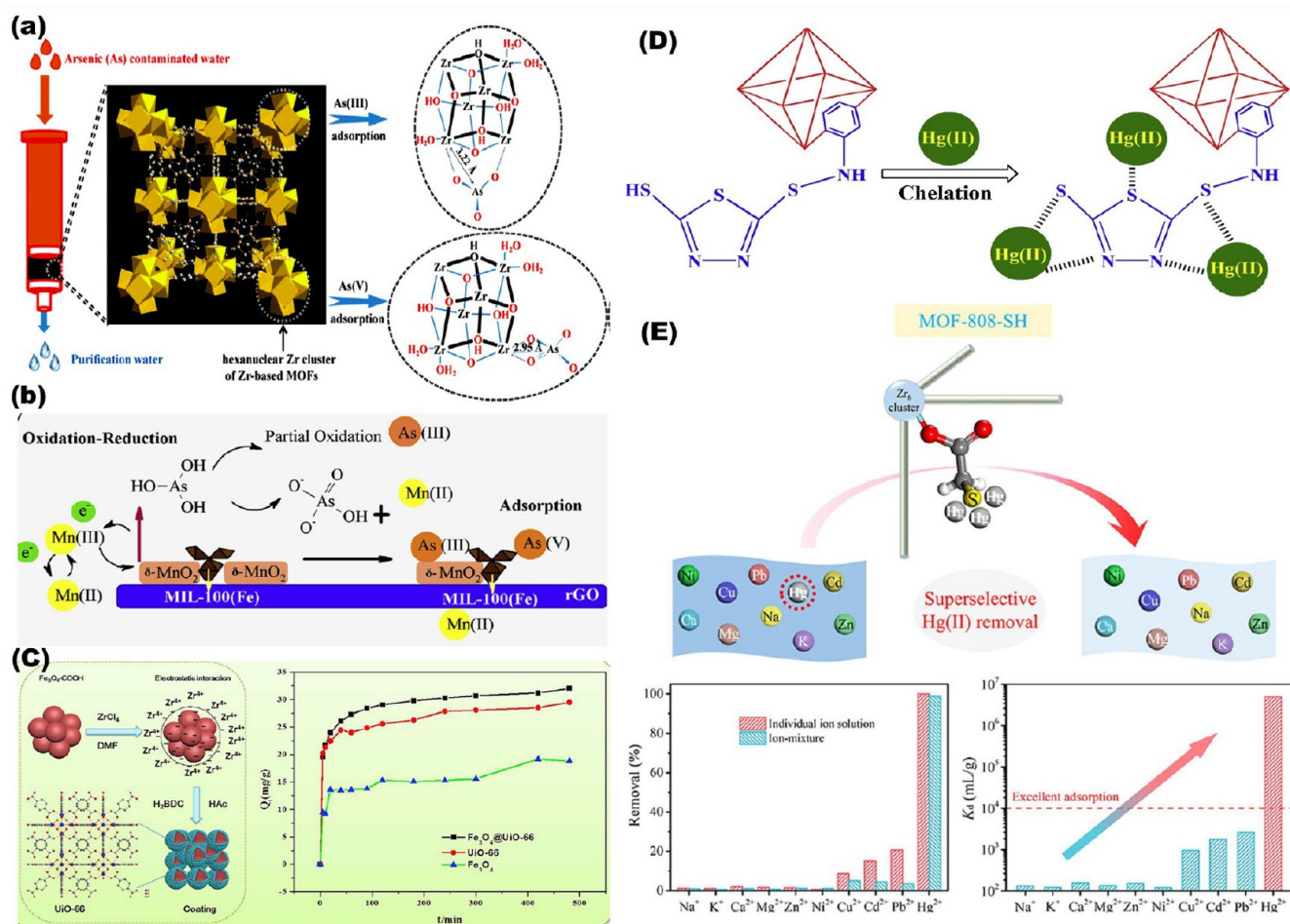
Figure 8. (a) General schematic representation for the synthesis of zirconium metal−organic frameworks [UiO-66 and UiO-66(NH₂)] for As(III) and As(V). Reproduced with permission from ref 98. Copyright 2019 Elsevier. (b) Fe-based metal−organic framework (MOF) MIL-100 (Fe) was synthesized and employed in the adsorption of As(III) and As(V). Reproduced with permission from ref 99. Copyright 2020 Elsevier. (c) General adsorption mechanism of magnetic metal−organic frameworks Fe₃O₄@UiO-66 for disregarding arsenic from water. Reproduced with permission from ref 100. Copyright 2019 Elsevier. (d) Development of UiO-66-NH₂ with 2,5-dimercapto-1,3,4-thiadiazole. Reproduced with permission from ref 102. Copyright 2019 Elsevier. (e) MOF-808 was grafted using thioglycollic acid to create the MOF-808-SH adsorbent for the elimination of Hg(II). Reproduced with permission from ref 103. Copyright 2022 Elsevier.

cleanup. Because of their reversible adsorption capability, MOFs support sustainable water treatment solutions by facilitating regeneration and making them reusable.

Radioactive Metal Ions Sequestration. The radioactive isotopes known as radionuclides can pose major risks to human health and the environment.[75] To address the world's energy and environmental demands, nuclear energy is seen to be a viable alternative to fossil fuels.[76] However, there might be risks for humans if radioactive elements are unintentionally released into the environment.[77] Nuclear wastewater that contains a variety of radionuclides, with unreacted uranium consistently being one of the most important radioactive wastes.[78] A significant amount of nuclear waste streams also contains other typical radionuclides, including ⁹⁹Tc and ¹³⁷Cs, which are byproducts of nuclear fission between ²³⁵U and ²³⁹Pu.[79] Radionuclide absorption by MOFs is a significant area of environmental protection and nuclear waste management.[80] MOFs can be designed on the basis of their ability to selectively adsorb certain radionuclides, which can be achieved through functionalization or by the intrinsic properties of the MOF structure, thereby allowing them to target specific radionuclides while excluding nonradioactive elements.[81] The mechanism of

radionuclides adsorption using MOFs is often driven by complex coordination interactions.[82] MOFs can be applied in nuclear waste storage and remediation efforts to effectively immobilize radionuclides and prevent their release into the environment. Rigorous analytical techniques, such as gamma spectroscopy or liquid scintillation counting, are used to monitor and confirm the removal and sequestration of radionuclides using MOFs (see Table S3).

Polyoxometalates (POMs) were introduced by Zhang et al. to prepare MOFs, and the POM-based MOF was then investigated for adsorption of uranium.[83] This material exhibits abundant redox sites, photocatalytic activity, and a strong interaction toward U(VI). Furthermore, the MOF's performance in uranium uptake and elimination was exceptional. When exposed to visible light, its maximum uptake was found to be 728.34 mg g⁻¹ and it absorbed over 99% of the uranium from the solution within 120 min. Additionally, a significant amount of nuclear fission products (841 g ton⁻¹) were extracted from the solution. Moreover, Liu and co-workers generated UIO-66-AO using diaminomaleonitrile as an intermediate. They demonstrated that amidoxime groups (AO) could be grafted onto UIO-66 and used for uptake of U(VI) with 106 mg/g.[84] A comparable
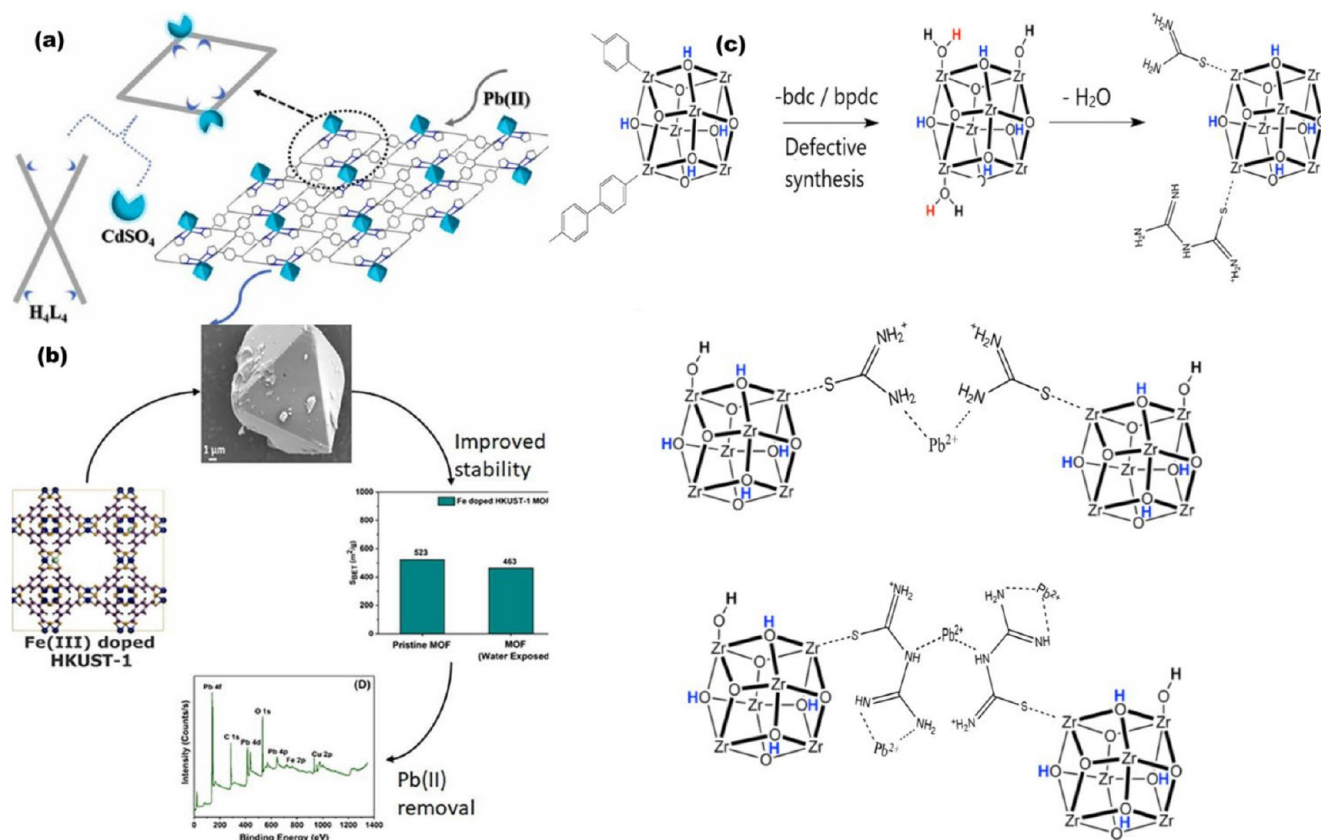
Figure 9. (a) Preparation of a three-dimensional metal−organic framework (Cd-MOF) by using $CdSO_4$ for the removal of Pb(II). Reproduced with permission from ref 106. Copyright 2022 Elsevier. (b) Schematic representation for Fe-enriched HKUST-1 MOF with different dopant concentrations for adsorption of Pb(II). Reproduced with permission from ref 107. Copyright 2022 Elsevier. (c) The engineering of zirconium MOFs functionalized with amines to remove lead(II) from water with thiourea and amidinothiourea were grafted onto pure UiO-66 and UiO-67 MOFs for removal of Pb(II). Reproduced with permission from ref 108. Copyright 2021 Elsevier.

procedure was employed to generate MIL-53 (Al)-AO in addition to UIO-66-AO for U(VI) adsorption with removal capacity around 100 mg/g.[85] Wu and co-workers employed the conventional solvothermal approach to manufacture the rodlike MOF-5, which was then utilized to extract U(VI) from radioactive wastewater.[86] At pH 5, the maximal U(VI) adsorption capacity onto MOF-5 was 237 mg/g. Furthermore, the introduction of $NaNO_3$ did not significantly reduce the adsorption capacity, thereby suggesting that MOF-5 might be an actual effective adsorbent in real applications (Figure 7a). According to the Liu group's studies,[87] MIL-101 (Cr) has a large specific surface area ($2482 \, m^2/g$) but a low U-uptake quantity at pH = 6 (91 mg/g). A postsynthesis approach was employed to add certain active functional groups to MIL-101 to improve its uranium adsorption capacity. To generate modified porous material (MIL-101-AO), they utilized a chloromethylated MOF MIL-101 combined with diaminomaleonitrile (DAMN) and amidoximation (Figure 7b). MIL-101-AO's outstanding selective adsorption capacity for U(VI) in aqueous solutions became achievable by the amidoxime group's chelation to uranium and broad specific surface area. Using a malononitrile intermediate, Mei and co-workers synthesized amidoxime-functionalized ZIF-90 (ZIF-90-AO).[88] ZIF-90-AO also showed a high capacity for adsorption of 468.3 mg/g at pH = 5 in a uranium adsorption experiment. ZIF-90, ZIF-90-OM, and ZIF-90-AO were regarded as outstanding candidates for the extraction of U(VI) from wastewater because of the aforementioned findings (Figure 7c).

Other Heavy Metals Elimination. One of the greatest significant environmental problems nowadays is heavy metal contamination, which is harmful to both humans and aquatic life.[89] The presence of toxic heavy metal ions, such as Hg(II), Pb(II), Cr(VI), As(III), As(V), and Cu(II), in surface water, primarily from the release of industrial effluent, is raising the risk of public health issues and endangering biological systems when tolerance limits are reached.[90] Several heavy metals, including lead (Pb), mercury (Hg), cadmium (Cd), chromium (Cr), copper (Cu), zinc (Zn), iron (Fe), manganese (Mn), and arsenic (As), have been associated with Alzheimer's disease (AD).[89−92] MOFs can be designed or selected on the basis of their affinity for certain heavy metals, and the choice may depend on factors such as ion size and charge.[93] The exceptional high surface area of MOFs could provide a large adsorption capacity when dealing with heavy metals present in water at low concentrations.[94] MOFs can be engineered to exhibit selective adsorption properties, which would allow them to bind specific heavy metal ions while ignoring other ions present in the water.[95] MOFs contain metal nodes or clusters connected by organic ligands, and some groups on the metal clusters can form coordination bonds with heavy metal ions[96] (see Table S4). After adsorbing heavy metals, MOFs can be treated to desorb metal ions or regenerate their adsorption capacity. After MOF treatment, heavy metal ion elimination from water is monitored and confirmed by using analytical approaches, such as atomic

absorption spectroscopy and inductively coupled plasma−mass spectrometry (ICP-MS).

Arsenic (As) poisoning is a serious worldwide problem because of its harmful impacts on human health and the ecosystem.[97] In order to eliminate arsenic (As) from polluted water, He and collaborators synthesized and studied zirconium MOFs [UiO-66 and UiO-66(NH$_2$)], which had removal capabilities of 205.0 and 68.21 mg/g for As(III) and As(V), respectively, as shown in Figure 8a.[98] As(V) and As(III) bidentate mononuclear complexes were mostly formed on the hexanuclear Zr cluster of UiO-66, as shown by the interaction mechanism. Furthermore, Fe-based MOF MIL-100 (Fe) was synthesized by Ploychompoo et al.[99] They embedded MIL-100 (Fe) with reduced graphene oxide (rGO) using a straightforward hydrothermal procedure, and it was coated with the manganese oxide via a one-pot reaction process to design MIL-100(Fe)/rGO/d-MnO$_2$ nanocomposites, which was applied for the removal of As. At pH 2, MIL-100(Fe)/rGO/d-MnO$_2$ nanocomposites showed adsorption capacities for As(III) and As(V) of 192.67 and 162.07 mg/g, respectively, and the saturated adsorbent was able to be regenerated for at least five runs (Figure 8b). Huo et al. fabricated magnetic MOF Fe$_3$O$_4$@ UiO-66 using a solvothermal method for eliminating arsenic from water and they determined the adsorption performance for arsenate to be a capacity of 73.2 mg g^{-1} (Figure 8c).[100]

Mercury ion is actually hazardous and persistent, and it poses a major risk to human health and the natural environment.[101] In order to eliminate the Hg(II) from water, Fu et al. developed a MOF adsorbent by postfunctionalizing UiO-66-NH$_2$ with 2,5-dimercapto-1,3,4-thiadiazole.[102] After studying the MOF's adsorption characteristics, they discovered that, at the ideal pH of 3, the highest adsorption quantity was 670.5 mg/g, and after 10 successive cycles, the adsorption efficiency was reduced by just 13.5%. According to their assumption (Figure 8d), the complexation process between Hg(II) and thiol/nitrogen-containing groups essentially drives the adsorption mechanism. By grafting thioglycollic acid onto MOF-808, Ji and co-workers created the MOF-808-SH adsorbent for the elimination of Hg(II).[103] The MOF-808-SH exhibited a distribution coefficient K_d of up to 5.0×10^6 mL/g, which indicated a high adsorption capacity of 977.5 mg/g. After six adsorption−regeneration cycles, MOF-808-SH showed recyclability with a minor loss of capacity. Furthermore, the ultrafast kinetics and high adsorption capacity for Hg(II) of MOF-808-SH may be explained by its large surface area, well-developed porous structures, and high SH loading (Figure 8e).

One of the main contaminants in water, lead, has detrimental effects on both the environment and the health of humans.[104] Lead exposure may adversely impact the central nervous system, decrease brain function, and damage the kidneys, liver, and reproductive system.[105] Wang et al. created a three-dimensional MOF (Cd-MOF) by using CdSO$_4$ and a multiple pyrazole heterocycle tetraphenylethylene-based ligand (H$_4$L$_4$) that contains Pb(II) adsorption sites (Figure 9a).[106] The Cd-MOF has stability and adsorption capacity for removal of Pb(II) (845.55 mg/g). Goyal et al. synthesized Fe-enriched HKUST-1 with different dopant concentrations of Fe (5−20 mol %) using a one-pot solvothermal technique (Figure 9b). Fe-doped HKUST-1 MOF demonstrated a Pb(II) adsorption capacity of 565 mg g^{-1} with removal efficiency > 90%.[107] Additionally, Morcos et al. constructed zirconium MOFs functionalized with amines to remove lead(II) from water via a simple one-step postsynthesis.[108] Thiourea and amidinothiourea were grafted

onto pure UiO-66 and UiO-67 MOFs using a simple one-step postsynthesis modification. For the removal of Pb(II), a process including dehydration, ionization, and coordination stages was described with high maximal adsorption capacities of 246 and 367 mg g^{-1}, respectively. They also explained the removal efficiencies to be over 95% with Pb (II) concentrations (25−250 ppm), and the MOFs could be regenerated for up to four cycles (Figure 9c).

■ CONCLUSIONS AND PERSPECTIVES

Advanced porous materials have a great deal of opportunity to influence the removal of a wide range of contaminants that exist in water sources because of their established pore architectures and tailored functionality. One of the greatest environmental challenges facing humanity is the effective removal of hazardous substances and water purification, which directly affects the well-being of every human being, as well as the health of our planet's ecosystem. MOFs have demonstrated potential in contributing to water purification because of their superior qualities in sorption applications, which include rapid sorption kinetics, great absorption capacity, and high selectivity. Despite significant developments in MOF research, a wide range of unexplored possibilities can be developed, which will continue to improve the performance of associated substances when these areas are revealed by future investigations. To fully explore this exciting potential in water purification, a few recommendations for overcoming the present challenges that are delaying their practical applicability are offered as follows:

(1) The complexity of transferring and processing bulk material is considered one of the major challenges. Therefore, to increase the amount produced, more effective approaches to synthesize and scale up MOFs are required. Currently, the process of producing MOFs generally uses a lot of resources and emits a lot of greenhouse gas emissions. Moreover, the synthesis of these task-specific MOF-based adsorbents often requires time-consuming synthetic methods, which makes their practical use even more difficult. To solve this issue, future research should concentrate on developing low-cost MOFs utilizing accessible, eco-friendly raw materials and improved synthesis approaches. This means that to fulfill industrial demands and guarantee the success of deploying these innovative materials outside of the lab, large-scale sustainable manufacture of MOFs-based products is needed.

(2) The certification requirements, which may be related to applications involving drinking water, present an additional obstacle to the widespread use of MOFs. To guarantee that a potential product will not have unjustifiably harmful impacts on people or the environment, the registration procedure often involves a thorough assessment of the product's chemical composition, consumption, hazards, and beneficial effects on society. MOFs cannot be introduced into the market unless they have been approved and identified by professional associations; alternatively, the public will not be sufficiently confident to use such materials to purify their drinking water. Additionally, different countries typically have varying registration procedures, which makes it more difficult to promote MOFs for real-world applications.

(3) The engineering of new MOFs as adsorbents is based on research that extends from the molecular to the macroscopic level. The direct relationship between pollutants and binding sites should be investigated to enhance the adsorbents efficiency. In addition, exhaustive analysis of the connection between sorption efficiency and macroscopic morphologies is also necessary.

Future research might focus on developing MOFs with more selectivity and efficiency for specific pollutants prevalent in water sources. The experimental directions may involve systematically modifying the structure and content of MOFs to improve their effectiveness against certain contaminants. Additionally, investigating innovative synthesis methods and postsynthetic modifications could help in the production of highly tailored MOFs. To evaluate the long-term stability of MOFs, researchers can perform long-term exposure studies that simulate a wide range of environmental conditions, such as fluctuating pH levels, temperature variations, and exposure to differing water chemistry. This might give useful insights into the long-term durability of MOFs and help to recommend strategies for enhancing stability. For optimal manufacturing procedures, researchers should explore different precursors, solvents, and reactor designs. Comparative research on the scalability of various synthesis pathways, including continuous flow processes, might provide useful information for enhancing the productivity of MOFs productivity. Furthermore, exploring the synergies between MOFs and other sophisticated water treatment technologies, such as membrane filtration or electrochemical processes, may improve the overall system performance. Researchers can develop procedures to determine the compatibility and effectiveness of incorporating MOFs into existing water treatment systems. Exploring hybrid systems and evaluating their performance under various conditions might lead to practical applications. Future studies should consider the environmental effects of MOF production and application. This information is critical for resolving environmental problems and satisfying regulatory obligations. Advanced characterization approaches, including in situ spectroscopy and imaging, can help offer a better understanding of MOF behavior during water treatment operations. Researchers may create experiments to monitor changes in the structure, identify active areas, and follow the outcome of pollutants in MOFs. This degree of information can help to build MOFs with better catalytic and adsorption characteristics. Collaboration among interdisciplinary research teams, including experts in materials science, environmental engineering, and chemistry, will be critical to overcoming these issues and broadening the practical uses of MOFs.

In conclusion, despite the recent progress of developing MOFs for water purification, the use of MOFs for water purification in real-world water bodies is a greater challenge than in a laboratory setting because of the complicated environment and the complex combination of different components. Therefore, future research should focus on boosting the chemical/hydrolytic stability, as well as durability and reusability, of MOFs that are cost-effective while developing scale-up and fabrication processes for their deployment in practice.

■ ASSOCIATED CONTENT

ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.langmuir.3c03818.

> A listing of tables, various types of MOFs with different adsorption properties for the removal of organic dye, oil sorption performance, and heavy metals sorption and their maximum adsorption capacity (PDF)

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Notes

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