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Mercury in aquatic environments: toxicity and advances in remediation using covalent organic frameworks

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Mercury is a dangerous and toxic contaminant that poses significant threats to both the environment and human health. Consequently, lowering mercury levels to meet drinking water standards has become a priority. Covalent organic frameworks (COFs) are porous materials with adjustable structures that are easy to design and synthesize, and they have garnered significant attention. COFs present a crucial solution for the urgent removal of mercury from water due to their outstanding properties, including high adsorption capacity, excellent photocatalytic performance, tunable porosity, and high stability and reusability. This review aims to discuss the latest advancements regarding the toxicity of mercury and its chemical forms, as well as the synthesis methods for COFs. For the first time, we introduce COFs and their applications in mercury removal, detailing the current challenges and issues faced by COF materials in this area. Additionally, we propose future research directions and obstacles to be addressed.

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1 Introduction

The rapid economic and social development has resulted in significant environmental pollution, leading to the generation of large quantities of toxic and hazardous waste.[1–3] These substances are released into water systems, creating sewage and posing serious health risks.[4] As a result, the removal of toxic waste from wastewater has become vital. Sewage mainly comprises heavy metals and organic pollutants.[5–7] Although microorganisms have been successfully utilized to treat organic pollutants *via* bioelectrochemical methods, heavy metals such as mercury, cadmium, chromium, lead, and arsenic cannot be broken down biologically.[8] These toxic heavy metals interact with proteins and enzymes in the body, causing them to become inactive and potentially resulting in chronic poisoning as they accumulate in specific organs.[9] Therefore, it is essential to remove heavy metals from water.

Mercury (Hg) is recognized as the third most toxic element to human health by the U.S. Agency for Toxic Substances and Disease Registry (ATSDR). Pure Earth estimates that mercury exposure poses health risks to 19 million people globally, with a disease burden of approximately 1.5 million Disability-Adjusted Life Years (DALYs). In 2015, the Toxic Sites Identification Program reported around 450 sites worldwide where mercury exposure jeopardizes public health.[10] While environmental mercury contamination is a critical global health issue, the complexity of mercury's biogeochemical cycle complicates precise assessments of its environmental and human health impacts.[11] Human activities have almost tripled the levels of mercury in the atmosphere, with its atmospheric concentration increasing by 1.5% annually. Mercury-contaminated soil or the movement of polluted water can enter the food chain through plants and livestock.[12]

The removal of mercury in various forms such as elemental, ionic, and organometallic (such as methylmercury) have garnered significant research attention due to its harmful and hazardous effects on ecosystems.[13] Mercury accumulating in environmental water often settles into sediments and transforms into toxic methylmercury. This methylmercury then enters the food chain, leading to severe diseases in living organisms.[14] Inorganic mercury can undergo biotic methylation to form dimethyl mercury, its most toxic variant, which can lead to nervous system disorders and kidney and liver damage, and hinder childhood development.[15] Consequently, the United States Environmental Protection Agency and the World Health Organization have set the maximum permissible levels of mercury in drinking water at 0.001 mg L^{-1} and 0.002 mg L^{-1}, respectively.[16] To address this, various remediation methods are being employed to remove Hg^{2+} from water.

Typically, conventional techniques like reverse osmosis,[17] adsorption,[18–20] photocatalysis,[21] ion exchange,[22] electrochemical treatment,[23] and membrane filtration[24] are employed to eliminate

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toxic mercury from water. However, it is important to note that the photocatalytic removal of heavy metals often faces limitations due to reduced light transmittance caused by scattering effects, which diminish photocatalytic efficiency.[25] Adsorption is favored over other methods due to its high efficiency, simplicity, and cost-effectiveness.[26] Porous materials, in particular, have gained significant attention for mercury adsorption due to their rapid diffusion rates and efficient mass transport of mercury ions. Various porous materials, including clays,[27] hydrogels,[28] zeolites,[29] silica gel,[30] activated carbon,[31] porous organic polymers,[32] and metal–organic frameworks (MOFs),[33–36] have been developed and utilized for this purpose. Nevertheless, the practical application of these adsorbents is often limited by challenges such as insufficient chemical stability, poor performance in highly acidic environments, low adsorption capacity, the lack of reusability, and poor selectivity.[37] COFs are crystalline materials with well-organized porous structures, created through thermodynamically controlled reversible polymerization of light elements *via* covalent bonds.[38–42] Unlike traditional materials, COFs are characterized by their low density, excellent chemical stability, uniform pore structures, high specific surface area, ease of functional design, and, in some cases, unique luminescence properties.[43] Hence, it is crucial to design COFs with enhanced catalytic activity and adsorption capacity for effective mercury removal from water.

In recent years, increased attention has been directed toward COFs, as evidenced by the growing number of papers published annually (Fig. 1). Researchers have focused on using COFs to remove toxic mercury, with approximately 42 publications on this topic steadily increasing from 2016 to the present. Considering the significance of environmental remediation and the considerable potential of COFs as versatile functional materials in this area, in this review, we thoroughly investigate the latest developments in using COFs for the removal of mercury from water. We start by highlighting the necessity of eliminating toxic pollutants and examining the properties of mercury and its toxic effects on human health. Next, we explore COFs, including their synthesis and characteristics. We then discuss the mechanisms involved in mercury removal in detail. Additionally, the article critically evaluates recent advancements in reducing mercury levels in water. Finally, we address the limitations and future potential of COFs in the removal of heavy metals.

2 Principle of mercury and its toxicity

2.1 Forms of mercury

Various environmental sources contribute to different chemical forms of mercury, including elemental mercury (metallic), inorganic mercury, and organic mercury. Elemental mercury (Hg^0) is released from items like thermostats, thermometers, dental amalgams, and latex paint, partly entering the atmosphere as vapor. As the only metal in a liquid state at room temperature, Hg^0 plays a significant role in both global mercury cycling and occupational health risks. It is easily absorbed through inhalation and can cross the blood–brain barrier, where it is rapidly oxidized to ionic mercury inside cells, remaining in brain tissues for years. In many regions, dentists continue to use dental amalgams, which primarily consist of elemental mercury.[44] Inorganic mercury (mercury salts) has been identified in products such as laxatives, cosmetics, teething powders, diuretics, and antiseptics. It can also form as a result of the metabolism of elemental mercury vapor or methylmercury (MeHg).[45]

Compounds formed from the combination of hydrogen and carbon are known as organic mercury compounds, which are categorized into allylmercury and alkylmercury compounds. Allylmercury compounds include phenylacetate mercury, used in agriculture, and mercurochrome, an antiseptic. Alkylmercury compounds consist of methylmercury and ethylmercury. Monomethylmercury and dimethylmercury, often found in ecological systems, can significantly contaminate marine environments. A well-known example is Minamata disease in Japan, caused by consuming fish and shellfish contaminated with mercury.[46] MeHg is the most prevalent form of organic mercury and a primary source of organic mercury in ecosystems. It is readily carried by water into aquatic systems and, due to its low water solubility, is considered relatively lipid-soluble. MeHg is easily absorbed by lower organisms, moves up the food chain, and bioaccumulates in fish. Consequently, fish are the main source of MeHg exposure and poisoning in humans.[47] Mercury pollution

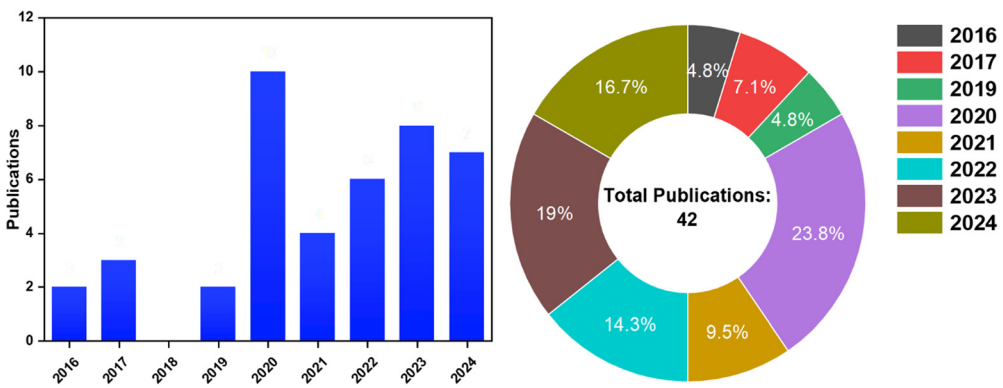

Fig. 1 The number of COF publications from 2016 to 2024 was analyzed. The Web of Science database was queried using the search terms 'COF' or 'Covalent–Organic Framework' along with 'mercury removal' or 'mercury capturing,' which were required to appear in the abstract.

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has become a global concern, drawing significant attention from the international community for three primary reasons. First, mercury has high long-range mobility. Its volatility and ability to disperse in the atmosphere allow it to travel far from its source, even reaching remote areas like the Arctic. Second, mercury is highly persistent, as it does not naturally break down in the environment. Third, methylmercury undergoes biological accumulation, making it challenging to eliminate once it enters living organisms. As humans occupy the top of the food chain, they face the greatest risk from mercury pollution.[48]

2.2 Mercury in the environment

Human activities have released significant amounts of anthropogenic Hg into aquatic environments, severely impacting environmental and ecological health on multiple levels.[49] Mercury pollution can arise from both natural processes and improper discharges, with the latter being the primary contributor due to rapid industrialization and high energy consumption in recent years. Key sources include lead-acid battery production, coal and fossil fuel combustion, mining, metal smelting, and waste incineration.[50,51] Excessive Hg enters waterways through pathways such as atmospheric deposition, runoff from catchments, and industrial or urban discharges. The U.S. EPA identifies mercury as a priority pollutant due to its persistence, high toxicity, and inability to biodegrade. Exposure to mercury contamination poses significant risks to human health, with the World Health Organization (WHO, 2006) setting a stringent safety limit of less than 0.006 mg L^{-1} for mercury in drinking water.[52]

Monitoring of surface water has revealed that mercury pollution is the predominant heavy metal contamination in China, along with other heavy metals such as lead, chromium, cadmium, and arsenic. Mercury ions are the most toxic and can cause the death of organisms even at low concentrations.[53] The Minamata disease outbreak in Japan, caused by mercury chloride released from chemical plants into seawater, is a stark reminder of its devastating effects. Mercury ions are particularly stable when bound to sulfur ions. Since proteins in the human body contain many sulfur-containing groups, mercury readily binds to thioredoxin reductase in the brain, deactivating the protein and leading to brain death.[54]

Hg^{2+} poses a significant threat to human health because inorganic mercury can be biochemically transformed into methylmercury, drastically increasing its toxicity. In contrast, arsenic ions undergo methylation to form the less toxic dimethylarsenic. The presence of a methyl group in methylmercury enhances its fat solubility, making it highly absorbable by the human body, with an absorption rate of up to 100%.[55] This property contributed to the tragic death of scientist Karen Wetterhahn, who accidentally spilled methylmercury on her hand. Alarmingly, any form of mercury in the environment can potentially convert into methylmercury under specific conditions, amplifying its toxicity. Thus, removing mercury ions before they transform into methylmercury is crucial for preventing serious health risks.[56] Various sources of mercury are presented in Fig. 2.

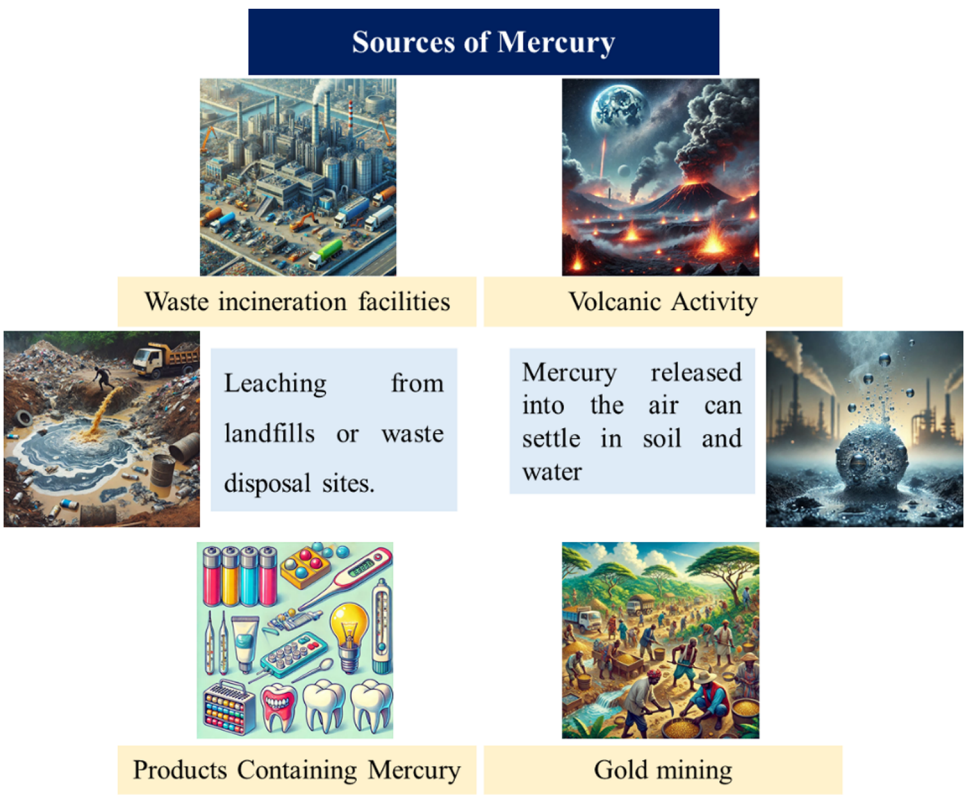

Fig. 2 Sources of mercury.

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2.3 Mercury toxicity on human health

Mercury in the environment originates from both natural and human-made sources. Major natural contributors include volcanic eruptions, rock weathering, forest fires, and the evaporation of mercury from soil and water. These processes release mercury that has accumulated from both natural activities and anthropogenic emissions. Mercury is a potential agent of harmful effects on humans, particularly as it is commonly found in occupational settings.

The greatest health risk associated with mercury arises from human exposure to MeHg through dietary sources, primarily aquatic animals such as fish. MeHg enters the human body *via* ingestion and is distributed to tissues through bloodstream absorption.[57] It primarily accumulates in the brain (central nervous system), kidneys, and liver. High levels of mercury exposure can lead to severe health effects, including hearing loss, vision problems, speech difficulties, neuronal cell death, and potentially fatal conditions.[58] Additionally, MeHg crosses the placenta, negatively impacting fetal brain development, making prenatal life more vulnerable to mercury toxicity than adulthood. The severity of Hg toxicity depends on the dose and rate of exposure to different mercury forms, with the brain being the main target for inhaled mercury vapor. Human exposure to mercury is assessed by measuring its levels in hair, urine, and blood.[59,60]

Neurotoxicity refers to any harmful impact on the central nervous system (comprising the brain and spinal cord) or the peripheral nervous system (which includes nerves and ganglia outside the central nervous system (CNS) that are affected by exposure to toxic substances).[61] Numerous studies on the effects of mercury exposure have underscored its classification as the most neurotoxic agent. High levels of mercury exposure can result in severe neurological damage and fatalities.[62] Chronic exposure has been linked to various neurological issues, including visual field defects, cerebral palsy, and deafness, as well as neuromotor problems such as ataxia, muscle weakness, numbness in limbs, chewing difficulties, tremors, and spasticity.[63] Cardiotoxicity primarily refers to damage to the heart muscle, resulting in impaired heart function. In the last decade, the effects of mercury on cardiac tissue have become increasingly recognized.[64] Mercury exposure is closely linked to conditions such as atherosclerosis, heart disease, stroke, heart attack, high blood pressure, carotid artery obstruction, and irregular heart rhythms.[65]

Mercury has long been recognized for its detrimental effects on immune system function, likely due to its negative impact on polymorphonuclear leukocytes (PMNs). Mercury inhibits PMN activity by diminishing their ability to eliminate foreign substances and suppressing the synthesis of adrenocorticosteroids, which hinders proper PMN formation.[66] Different species of mercury, including methylmercury and inorganic mercury, have been found to induce genotoxicity in humans through the production of free radicals, leading to oxidative stress, disruption of microtubules, and interference with DNA repair processes.[67] Genotoxic effects on chromosomes and DNA disruption have been observed in individuals exposed to high levels of mercury.[68] Mercury is also considered a potential carcinogenic agent, particularly in occupational settings. Substantial evidence shows a significant correlation between occupational exposure to mercury (as measured in toenails, hair, and blood) and an increased risk of cancer and mortality,[69] as depicted in Fig. 3.

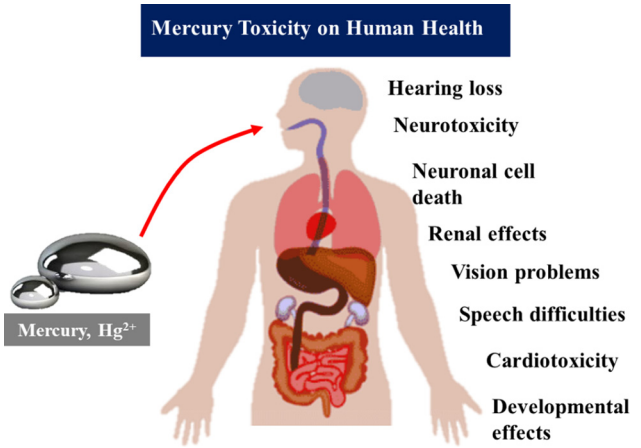

Fig. 3 Toxic effects of mercury on human health.

3 Covalent organic frameworks and their synthetic methods

Reticular chemistry, which involves connecting molecular building blocks through strong covalent bonds to form crystalline extended structures, has led to the development of several new classes of porous materials, with COFs being the most recent example.[70] Fundamentally, the directional nature of covalent bonds enables precise control over how building units assemble into predesigned structures. This characteristic, combined with the strength of the linkages, results in robust materials with significant potential for various applications.[71] COFs are recognized as a new class of crystalline framework materials and are often regarded as counterparts to MOFs, following the groundbreaking work reported by Yaghi *et al.* in 2005.[72] These frameworks are constructed entirely from organic building blocks, guided by the principles of reticular chemistry.[73] COFs possess tunable molecular secondary structural units, allowing the generation of various configurations within a periodic and well-defined architecture. Fig. 4 illustrates several building units that have been successfully utilized in the synthesis of COFs. The symmetry, size, and connectivity of the linkers determine the geometry of the resulting framework.

3.1 Solvothermal synthesis

The initial synthesis of COFs was carried out using a solvothermal reaction, which remains the most widely used and favored method for COF production. In this approach, COF precursors are combined in a vial with the chosen solvent or solvent mixture, along with catalysts or modulators if necessary. The reaction vessel must be a sealed container, for example, a Pyrex tube, a steel autoclave with a Teflon lining, or a glass bottle with

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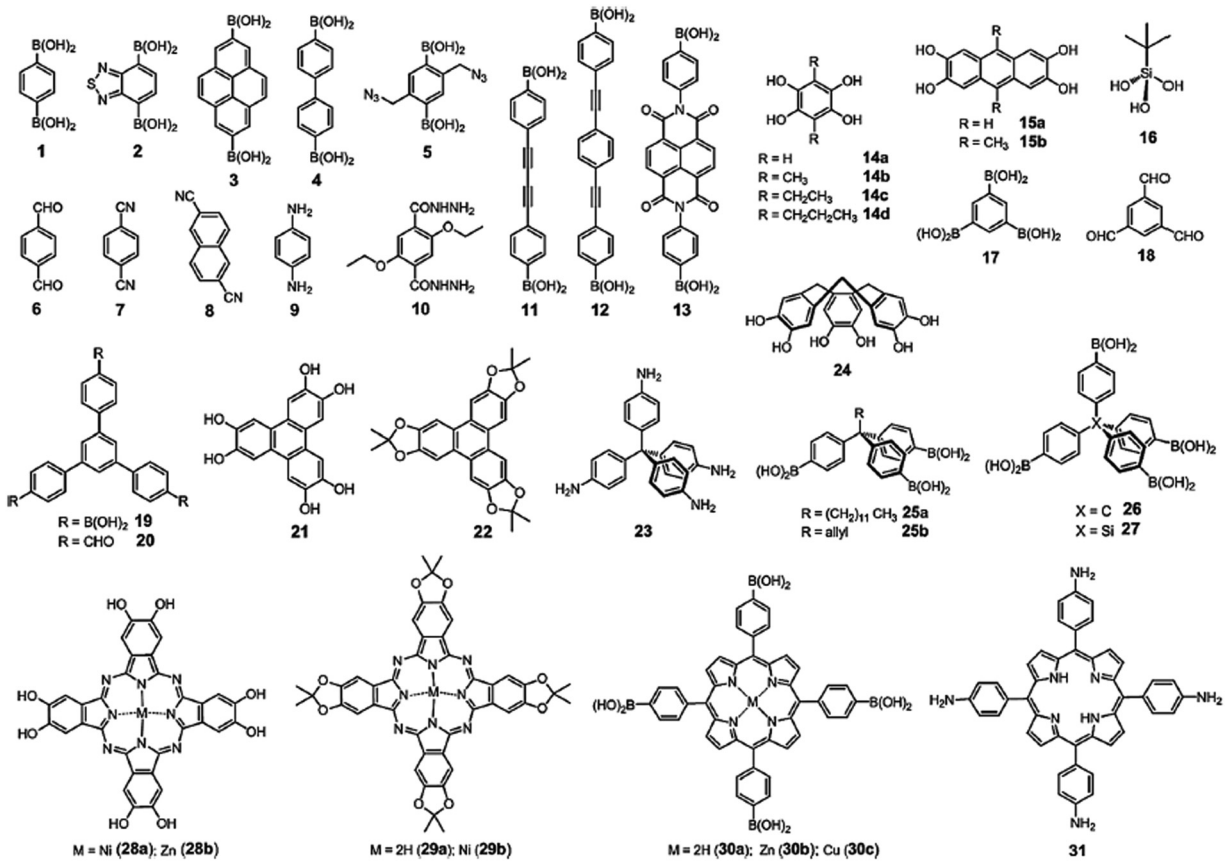

Fig. 4 Building units successfully utilized for the synthesis of COFs.[74] Copyright 2013, The Royal Society of Chemistry.

a plastic cap. This setup allows the reaction mixture to be heated above the solvent's normal boiling point, enhancing the solubility of the precursors and improving reaction kinetics.[75] Laemont *et al.* demonstrated that achieving high-quality COFs required the use of an apolar solvent combined with precise amounts of water and acetic acid. Under consistent reaction conditions, they successfully synthesized fifteen different COFs using an environmentally friendly solvent mixture of *n*-butanol, acetic acid, and water at 70 °C with continuous stirring for 16 hours. The resulting COFs exhibited crystallinity comparable or even superior to those produced *via* conventional solvothermal synthesis. Furthermore, this method proved highly scalable, enabling the synthesis of over ten grams of COF in a single batch.[76]

3.2 Ionothermal synthesis

Ionic liquids are generally defined as salts primarily consisting of ionic components, with melting points below 100 °C.[77] Their ionic nature imparts distinctive physicochemical properties, including exceptionally low vapor pressure and high thermal stability. As a result, ionic liquids have been widely studied in areas such as extraction, synthesis, and catalysis.[78] Compared to traditional organic solvents, ionic liquids are recognized as environmentally friendly and safe reaction media for chemical processes. Ionic liquids have recently garnered significant attention as environmentally friendly and safe reaction media for synthesizing crystalline materials, such as zeolites, MOFs, and COFs. This interest stems from their unique properties, including negligible vapor pressure, nonflammability, a broad liquid range, excellent solubility for both organic and inorganic compounds, and highly customizable structures.[79]

3.3 Sonochemical synthesis

High-intensity ultrasound has proven highly effective in the synthesis of organic, organometallic, and various advanced materials, emerging as a green alternative technology. In sonochemistry, acoustic cavitation resulting from the formation, growth, and implosive collapse of bubbles in a liquid due to ultrasonic shearing plays a central role. This phenomenon generates localized hot spots, which significantly accelerate chemical reactions.[80] During sonochemical synthesis, the crystallization process is enhanced as the high pressure and temperature produced by acoustic cavitation in the solution increase both cooling and heating rates. Additionally, this method is energy-efficient and cost-effective, as it eliminates the need for an induction period.[81] Wei *et al.* rapidly synthesized a new COF using the sonochemical method at room temperature in just 60 minutes, eliminating the need for toxic reagents. Following this, adsorption experiments demonstrated the material's outstanding adsorption capacity and selectivity for flavonoids. The sonochemically synthesized COF displays favorable characteristics, including high adsorption capacity,

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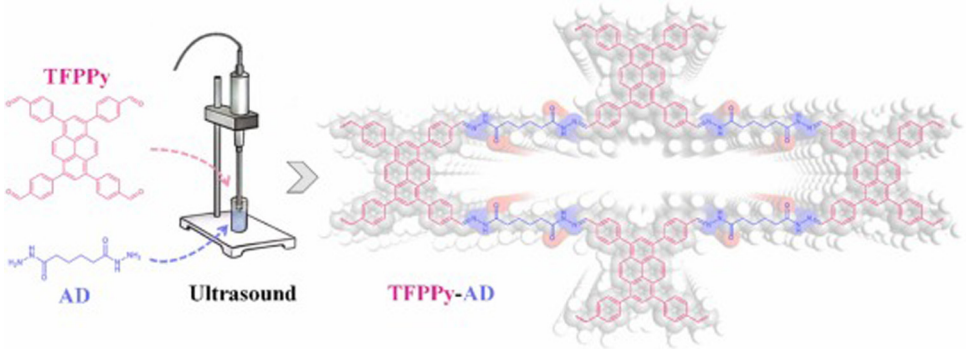

Fig. 5 Synthesis of the COF.[80] Copyright 2024, Elsevier.

selectivity, and a quick adsorption rate for flavonoids,[80] as shown in Fig. 5.

3.4 Mechanochemical synthesis

Efficient, simple, and environmentally friendly methods for synthesizing porous COFs are highly desirable, particularly for industrial-scale production. Mechanochemical synthesis, though an established technology, presents a promising approach for large-scale COF manufacturing.[82] During the ball milling process, solid raw materials are subjected to dynamic impacts from moving balls, leading to particle size reduction, shape alteration, bond breaking, and the creation of new surfaces at room temperature through the kinetic energy of the balls.[83] Compared to solvothermal synthesis, ball milling offers potential advantages such as scalability, energy efficiency, and cost-effectiveness. Additionally, its ability to minimize or eliminate solvent use makes it an increasingly appealing method.[84]

3.5 Microwave synthesis

Microwave heating is a widely used technique for accelerating chemical reactions. Studies have shown that crystalline MOF materials can be synthesized using this method.[74] In 2008, Cooper and his team introduced a rapid method for synthesizing COF materials using microwave-assisted techniques.[85] Microwave heating enables a cleaner and faster synthetic process compared to the solvothermal method, offering new opportunities for large-scale applications. In 2009, Cooper's group pioneered the microwave-assisted synthesis of boronate ester-linked COFs (COF-5 and COF-102). Under microwave irradiation at 100 °C, COF-5 rapidly formed within 20 minutes through the condensation of 1,4-benzenediboronic acid and 2,3,6,7,10,11-hexahydroxytriphenylene (HHTP) in both sealed and open containers—an impressive 200 times faster than traditional solvothermal synthesis. Notably, the resulting COF-5 exhibited a significantly higher Brunauer–Emmett–Teller (BET) surface area (2019 $m^2 g^{-1}$) compared to its solvothermal counterpart (1590 $m^2 g^{-1}$), attributed to the efficient removal of trapped impurities *via* an advanced microwave extraction process,[42] as shown in Fig. 6.

3.6 Plasma-induced synthesis

Plasma is commonly known as the "fourth state" of matter. Plasma discharges, consisting of free electrons, radicals,

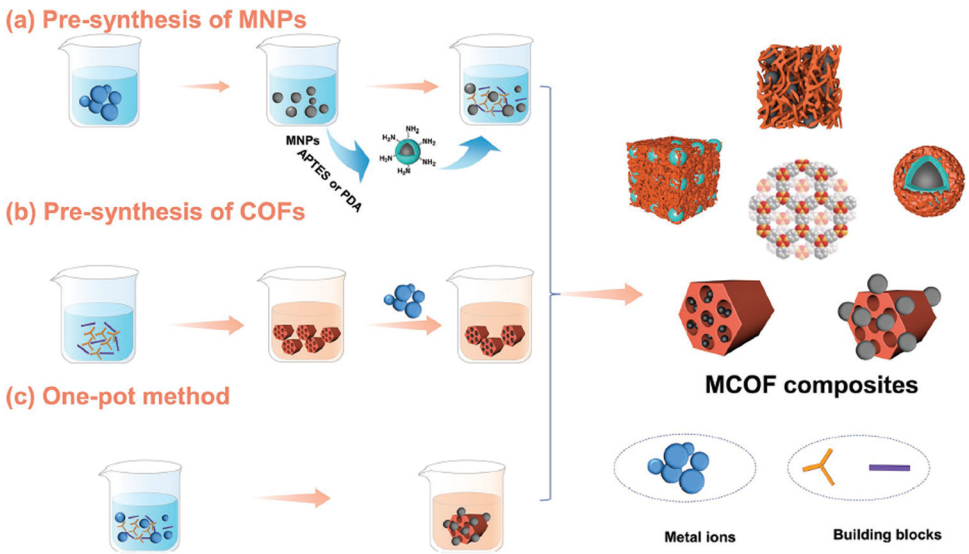

Fig. 6 The general preparation procedures of magnetic COFs.[42] Copyright 2023, Wiley.

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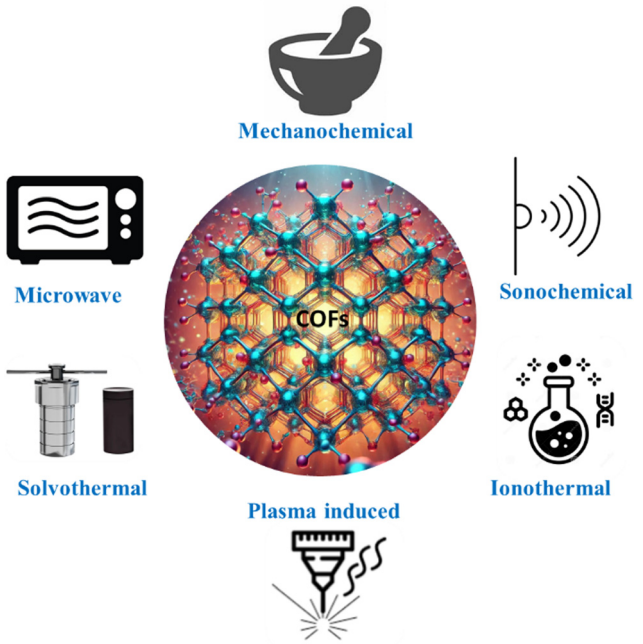

Fig. 7 Synthesis methods for COFs.

photons, metastable species, and excited neutral particles, can be utilized to initiate chemical reactions. Notably, non-thermal plasmas, which produce reactive species at atmospheric pressure and near room temperature, have found extensive applications in the synthesis of nanomaterials and polymers.[86] Qing *et al.* noted that COFs are usually synthesized under solvothermal conditions involving high temperatures and extended reaction times ($\geq$120 °C, >72 hours). In this study, they introduce a rapid microplasma electrochemical strategy for synthesizing COFs under ambient conditions. This method enabled the preparation of a series of flexible, high-crystallinity imine-bond COFs in just a few minutes, achieving a space-time yield that is 1000 times greater than that of the traditional solvothermal approach. Additionally, the method facilitated the synthesis of COFs with various linkages, including rigid imine, hydrazone, β-ketoenamine, and azine linkages. Notably, four types of imine-based COFs were synthesized in aqueous acetic acid, eliminating the need for harmful organic solvents, which demonstrates that the microplasma method is both environmentally friendly and versatile for COF synthesis.[87] All synthesis methods for COFs are shown in Fig. 7.

The advantages and disadvantages of the synthesis strategies for COFs are summarized in Table 1.[88,89]

4 Mercury removal by COFs

Some COFs are engineered with coordinating groups embedded in their walls, enabling them to capture metal ions. In addition to their high surface areas, COFs can be regarded as nanostructured coordinating macroligands with the added benefit of adjustable pore sizes. This tunability allows for selective ion capture by accommodating ions of varying radii, which can impact the adsorption equilibrium constant and be optimized for specific selectivity.[90] Recent research highlights the effectiveness of COFs as adsorbent materials, particularly for removing mercury ions from water.

The study by S. Archana *et al.*[91] investigates the ultrasound-assisted solvothermal synthesis of BDTA-COF, a COF derived from 4,4′-diaminobiphenyl and trialdehyde, for efficient Hg^{+} ion removal from aqueous solutions *via* adsorption. The material achieved a maximum adsorption efficiency of 99% through the optimization of parameters such as metal selectivity, concentration, pH, and adsorbent dosage. Nitrogen atoms in the imine linkages and oxygen atoms within the BDTA-COF were identified as the primary active sites for Hg^{+} adsorption. Kinetic studies indicated a pseudo-second-order chemisorption mechanism, while isotherm modeling followed the Langmuir model, suggesting monolayer adsorption. Rajeshkumar *et al.*[92] leveraged the unique properties of COFs for the efficient removal of Hg^{2+} ions. The COF nanosheets exhibited π–π transitions, effective electron transfer, and enhanced π-orbital delocalization within the BBT ring. Inductively coupled plasma (ICP) mass spectrometry analysis demonstrated that COF

Table 1 Advantages and disadvantages of COF synthesis strategies

Synthesis method	Advantages	Disadvantages
Solvothermal synthesis	• High crystallinity • A wide variety of solvents can be utilized • Good control over pore size	• Requires high temperatures and pressures • Cannot be used with insoluble building blocks • Solvent use can be problematic
Ionothermal synthesis	• Low volatility of ionic liquids • Can enhance the solubility of reactants	• High cost of ionic liquids • Requires high temperatures • Limited thermal stability
Sonochemical synthesis	• Rapid synthesis • Can produce nanoscale materials	• Equipment cost • Scale-up challenges • Applicable solely to small COFs
Mechanochemical synthesis	• Solvent-free process • Reduced environmental impact	• Often lower yields • Requires specialized equipment
Microwave synthesis	• Significantly reduced reaction time • Improved crystallinity • Synthesis at room temperature	• High equipment cost • Limited scalability for larger batches
Plasma-induced synthesis	• Can achieve high surface areas • Versatile in functionalization	• Complexity of plasma equipment • Process parameters can be difficult to control

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nanosheets removed nearly 99% of Hg^{2+}. Using coordination theory to exploit interactions between functional groups and Hg^{2+}, two covalent organic polymers were synthesized *via* a more environmentally friendly Michael addition–elimination reaction. A thiourea structure, known for its strong affinity toward Hg^{2+}, was successfully incorporated into the covalent organic polymer (COP) framework. This incorporation created a structure enriched with sulfur (S) and nitrogen (N) atoms, enabling high adsorption performance for mercury ions. Adsorption experiments revealed that the dithiourea-functionalized COPs achieved remarkable maximum adsorption capacities of 840.9 mg g^{-1} and 880 mg g^{-1} for the two materials, respectively. Even after four adsorption–desorption cycles, the materials maintained good adsorption efficiency and strong selectivity for Hg^{2+}.[93]

Zhu Ye *et al.* developed and synthesized three types of covalent organic polymers enriched with nitrogen, oxygen, and sulfur for the adsorption of mercury ions from solutions. To enable comparison, aldehyde building blocks containing one, two, or three thiophene rings were condensed with flexible hydrazide terephthalate, resulting in COPs designated as HTDP-1, HTDP-2, and HTDP-3. Among these, HTDP-2, featuring electron-rich thiophene-bicyclic systems, demonstrated rapid adsorption efficiency for Hg^{2+}, achieving 91.2% removal within just 10 minutes. This study reports the synthesis of a highly porous COF containing amine and sulfonyl functional groups using a hydrothermal method. The synthesized COF exhibited outstanding performance in the removal of Hg^{2+} ions, achieving an effective removal efficiency of 99.8% at neutral pH. Additionally, the COF was successfully applied as a probe for Hg^{2+} removal in natural river water samples.[94] Mohaddeseh *et al.*[95] focused on creating an adsorbent for removing mercury ions from aqueous solutions. They developed a porous covalent triazine ring-based nanoneedle using a straightforward hydrothermal synthesis method. The adsorbent's structure and morphology were analyzed using various characterization techniques, and its structural stability was evaluated under different temperature and pH conditions. The maximum adsorption capacity for Hg^{2+} ions was determined to be 1826 mg g^{-1} at ambient temperature and an optimal pH of 5. Adsorption data were analyzed using both linear and nonlinear models, with the linear Langmuir model showing an excellent fit (R^2 = 0.998).

Research studies on COFs for mercury removal, along with their parameters, are summarized in Table 2.

5 Mercury removal by functionalized COFs

COFs, with their regular and well-defined pore environments, can be functionalized during synthesis by incorporating various linkers to alter their pore characteristics. However, the range of possible pore environment modifications during synthesis is often quite limited. To overcome this constraint, post-synthetic modifications have been developed as an alternative strategy. These modifications enable the introduction of functional or large side groups, providing COFs with unique properties and significantly expanding their range of applications.[114] This review primarily focuses on advanced functionalized COFs, including those modified post-synthetically and COF-based composites. Additionally, it systematically highlights the applications of these functionalized COFs in mercury removal.

Hussain *et al.*[37] incorporated a desirable thiourea group into a flexible COF. Studies on the adsorption mechanism revealed that mercury adsorption preferentially occurred in the keto and thiol tautomeric forms rather than the enol and thione forms. COF showed exceptional mercury adsorption capacities (4270 and 4277 mg g^{-1}, respectively), attributed to their abundant chelating sites, with adsorption isotherms well-fitted to the Langmuir model. Additionally, both COFs exhibited remarkable stability under harsh conditions and achieved high mercury removal efficiencies (>97%) even in strongly acidic environments. Wang *et al.*[115] synthesized two thiol-modified

Table 2 Covalent organic frameworks used to remove mercury from water

Type of COF	Method	Catalyst dosage	Initial concentration	Temperature (°C)	pH	Performance (%)	Reaction time (min)	Recycling performance	Ref.
$COF_{DBD\text{-}BTA}$	Adsorption	0.33 g L^{-1}	30 μg L^{-1}	20	6.5	98	3	—	96
Thioether-based COF	Adsorption	5.0 mg	10.0 mg L^{-1}	—	5	99	—	—	97
Thiourea-linked COFs	Adsorption	5 mg	10.0 mg L^{-1}	25	4	99.7	0.17	4	98
COF	Adsorption	5.0 mg	10 ppm	—	7	99	20	6	99
Triarylamine-based COF	Adsorption	10 mg	10 μM	—	—	95	—	—	100
COF	Adsorption	25 mg	10 ppm	25	7	92	15	6	101
Magnetic COF	Adsorption	25 mg	20 mg L^{-1}	—	3	99	10	5	102
Azine-linked COF	Adsorption	10 mg	0.5 mmol L^{-1}	25	7	96.2	—	—	103
Sulfhydryl modified COF	Adsorption	0.1 g L^{-1}	1000 μg L^{-1}	25	7	99	10	10	104
β-Ketoenamine linked COF	Adsorption	10 mg	10 ppm	—	7	99.91	12	5	105
Pyridine-COF	Adsorption	2.0 mg	10 mg L^{-1}	20	5	96	—	5	106
Sulfur-rich COP	Adsorption	1 mg mL^{-1}	10 mg L^{-1}	25	3	99.9	5	—	107
$COF_{BTT\text{-}AMPD}$	Adsorption	15 mg	20 μM	—	—	—	20	—	108
Alkylamine COF	Adsorption	10 mg	10 mg L^{-1}	—	5	99.98	30	5	109
Triazole and methylthio modified COF	Adsorption	2.5 mg	0.5 mg L^{-1}	—	5–8	97.92	60	5	110
COF	Adsorption	5 mg	10 mg L^{-1}	—	6	96	30	5	111
Imine-linked COF	Adsorption	19.7 mg	10 mg L^{-1}	24	5.6	98.23	10	4	112
Alkynyl-based COF	Adsorption	25 mg	10 mg L^{-1}	25	6	99	10	5	113

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COFs: COF–S–SH and COF–OH–SH. These modified COFs exhibited excellent Hg(II) adsorption capacities, reaching 586.3 mg g^{-1} for COF–S–SH and 535.5 mg g^{-1} for COF–OH–SH. Both materials demonstrated outstanding selectivity for Hg^{2+} in the presence of various cationic metals in water. Interestingly, experimental results revealed a positive synergistic effect between co-existing toxic anionic diclofenac sodium and Hg^{2+} in enhancing the adsorption of each pollutant onto the COFs. Tao *et al.*[116] reported a SO_3^- anchored covalent organic framework ($[NH_4]^+[COF–SO_3^-]$) designed for mercury removal. This adsorbent demonstrated an ultrahigh Hg^{2+} adsorption capacity of up to 1299 mg g^{-1} and a remarkable uptake of 932.6 mg g^{-1} for Hg^0, making it one of the highest-performing porous solid adsorbents reported to date. The material exhibited excellent selectivity for Hg^{2+}, as evidenced by a high distribution coefficient of 2.3×10^5 mL g^{-1}. Breakthrough experiments further confirmed its high selectivity even in the presence of competing metal ions. Additionally, the adsorbent maintained its performance and structural integrity after four cycles, showing no significant loss in Hg^{2+} capture efficiency. Li *et al.*[117] developed a benzothiadiazole-based covalent organic framework (TPS-COF) through a one-step synthesis process. The material exhibited a high specific surface area of 1564 m^2 g^{-1}, excellent crystallinity, and stability, owing to its highly conjugated benzothiadiazole linkage structure. The presence of sulfur (S) and nitrogen (N) elements in the benzothiadiazole unit enabled outstanding Hg^{2+} adsorption performance, including a high adsorption capacity of 1040 mg g^{-1}, a rapid initial adsorption rate of 448 mg g^{-1} min^{-1}, and a short equilibrium time of just 10 minutes.

Lu *et al.*[118] investigated a carboxy-functionalized COF (COOH@COF) designed for the effective removal of Hg^{2+} from water. This COOH@COF demonstrates remarkable adsorption capacity for Hg^{2+} (99.1 mg g^{-1}) in aqueous solutions. The adsorption process follows the Langmuir isotherm model, while the kinetics adhere to a pseudo-second-order model. Notably, COOH@COF retains its adsorption capacity even after 20 cycles, highlighting its potential for practical applications in water purification. Yang *et al.*[119] developed three covalent triazine frameworks (CTFs)—MSCTF-1, MSCTF-2, and xSCTF-2—with varying pore sizes and S-group functionalization to enable selective removal of Hg^{2+} ions from aqueous solutions. Detailed structural analysis revealed that MSCTF-2, with a sulfur content of 24.45%, achieved the highest Hg^{2+} adsorption capacity of 840.5 mg g^{-1}. Meanwhile, MSCTF-1 demonstrated an exceptionally high distribution coefficient of 1.67×10^8 mL g^{-1}, effectively reducing Hg^{2+} concentrations in contaminated water to below 0.03 μg L^{-1}. The MSCTFs exhibited key advantages, including (i) high selectivity for Hg^{2+} over other transition metal ions, (ii) stability across a wide pH range,[1–12] and (iii) excellent recyclability, maintaining 94% Hg^{2+} removal efficiency over five cycles. Adsorption behavior followed pseudo-second-order kinetics and the Langmuir isotherm model. Wang *et al.*[120] developed a defect-rich Cu_2WS_4 nano-homojunction integrated with covalent organic frameworks (COFs) featuring numerous sulfur (S) vacancies. Through an ion pre-anchoring strategy, they achieved highly dispersed and uniform Cu_2WS_4 nanoparticles strongly immobilized on the COFs, resulting in significantly improved Hg^0 removal performance. The Cu_2WS_4@COF composites demonstrated a saturation adsorption capacity of 21.60 mg g^{-1}, which was nine times higher than that of Cu_2WS_4 crystals. Notably, these hybrid materials exhibited reduced adsorption deactivation at elevated temperatures, offering a broad operating temperature range (40–200 °C) due to the thermal stability of the active S species secured by both physical confinement and chemical interactions within the COFs.

Wang *et al.*[121] employed a tunable porous COF as a support for the *in situ* growth of silver nanoparticles (Ag NPs) using a one-step solution infiltration method. This approach enhanced the spatial dispersion of the nanoparticles, improved their stability in acidic solutions, and, for the first time, enabled the study of mercury adsorption performance. The composite demonstrated exceptional performance, including a high removal rate of 99%, an ultrahigh Ag atom utilization efficiency of 150%, excellent selectivity and stability, and reusability for mercury ion removal. Ge *et al.*[122] successfully prepared well-dispersed magnetic Fe_3O_4-decorated porous melamine-COFs. The resulting magnetic M-COFs featured large surface areas ranging from 344 to 600 m^2 g^{-1} and exhibited excellent magnetic separation capabilities (MS = 0.75–3.59 emu g^{-1}). Their adsorption properties were tested for the removal of heavy metal ions from wastewater. Notably, the synthesized Fe_3O_4/M-COFs demonstrated highly selective adsorption of Hg^{2+} ions with a high adsorption capacity of 97.65 mg g^{-1}. Yang *et al.*[123] developed a novel magnetic COF loaded with a ligand (MCM) that exhibited efficient adsorption capacity and a rapid removal rate for mercury ions. The MCM material was successfully utilized in a portable column unit to effectively separate mercury ions from real water samples, reducing mercury ion concentrations from 10.0 mg L^{-1} to 10.0 μg L^{-1} within just 15 seconds. Kinetic studies indicated that the adsorption behavior of MCM for mercury ions followed the pseudo-second-order model, while the adsorption equilibrium data aligned with the Langmuir model. Additionally, after five adsorption cycles, the MCM retained 96.89% of its initial adsorption capacity, demonstrating excellent regeneration potential.

Research studies on functionalized COFs for mercury removal, along with their parameters, are summarized in Table 3.

6 Conclusions and future perspectives

Inadequate global sanitation and the increasing need for wastewater reuse have intensified the prevalence of contaminants, posing significant risks to public health, the environment, and biodiversity. Current wastewater treatment methods are insufficient to fully address these challenges, necessitating collaborative efforts between industry and academia to develop innovative solutions. Among the contaminants of concern, mercury ions are particularly harmful due to their association with neurological disorders and cardiovascular effects, highlighting the critical need for their removal in both potable and non-potable water uses.

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Table 3 Functionalized covalent organic frameworks used to remove mercury from water

Type of COF	Method	Catalyst dosage	Initial concentration	Temperature (°C)	pH	Performance (%)	Reaction time (min)	Recycling performance	Ref.
Tb^{3+}-functionalized COF	Adsorption	100 mg L^{-1}	—	—	7	—	25	—	124
Vinyl-functionalized COF	Adsorption	5 mg	5 ppm	50	—	99.94	10	—	125
Thiol-functionalized COF	Adsorption	10 mg	800 μg mL^{-1}	25	3–4	94.1	50	5	126
Allyl with hydroxy group functionalized COF	Adsorption	20 mg	10 ppm	—	—	95	—	4	127
Thioether-functionalized COF	Adsorption	5 mg	10 mg L^{-1}	—	—	98	—	—	128
Bifunctional thiophene-based COFs	Adsorption	—	5 mg L^{-1}	—	7	99	—	5	129
Thioether-functionalized COF	Adsorption	10 mg	10 ppm	—	—	—	10	5	130
Fe_3O_4@RCOF-EBH	Adsorption	0.01 g/100 mL	10 mg L^{-1}	25	6	99.8	5	4	131
Thiol and thioether-functionalized magnetic COF	Adsorption	5 mg	—	—	6	95	10	5	132

Conventional water purification technologies often fail to eliminate toxic metals, with adsorption emerging as the most researched and applied method. Efforts have focused on creating low-cost, eco-friendly adsorbents, utilizing materials like silica, clay, zeolites, and activated carbon. Recently, COFs have gained prominence due to their high porosity, adjustable pore structures, and selective adsorption capabilities. Through rational monomer design, COFs exhibit strong affinity for metal ions *via* electrostatic interactions, π–π interactions, hydrogen bonding, and size exclusion. COFs have demonstrated significant potential for mercury ion removal, offering tailored nanomaterials for selective adsorption. However, challenges such as cost reduction and scaling-up production remain. As a relatively new field, COFs for water purification show immense promise, with further exploration of their structure–performance relationships expected to drive advancements in wastewater treatment technologies, surpassing traditional organic and inorganic materials.

The future of COFs lies in enhancing their structural stability, synthesis strategies, and functional applications. Research should focus on developing robust COFs with improved chemical and thermal stability, exploring green and scalable synthesis methods, and optimizing hierarchical pore structures to enhance adsorption and separation performance. COFs have promising applications in energy storage, catalysis, environmental remediation, and biomedical fields, but challenges such as biocompatibility, scalability, and long-term stability must be addressed. The integration of machine learning and computational simulations can accelerate the discovery of novel COFs with tailored properties. Additionally, industrial-scale production and commercialization will be key research areas for unlocking the full potential of COFs in real-world applications, including energy, healthcare, and environmental science.

Data availability

Data sharing is not applicable to this article as no datasets were generated or analysed during the current study.

Conflicts of interest

There are no conflicts to declare.

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