CELLULOSE BEADS, CELLULOSE-BASED METAL-ORGANIC FRAMEWORKS AND HETEROPOLYACIDS IN WATER TREATMENT: A REVIEW

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Water treatment is crucial for ensuring the availability of clean and safe drinking water, and the increasing prevalence of water contamination has driven the development of advanced functional materials for efficient purification. This critical review presents a comprehensive evaluation of cellulose-based materials, metal-organic frameworks (MOFs), and heteropolyacids (HPAs) for water treatment applications. Emphasis is placed on the preparation strategies of cellulose beads, cellulose-based MOFs (CelloMOFs), and HPAs, along with their structural features and functional properties. The performance of these materials in adsorption processes for removing heavy metals, organic pollutants, and dyes is systematically discussed. In addition, their photocatalytic and catalytic reduction degradation activities are examined, particularly their roles in advanced oxidation processes (AOPs). Overall, this review highlights recent progress, identifies current limitations, and outlines prospects for the development of efficient and sustainable water purification systems, with particular emphasis on preparation methods.

Keywords: cellulose, MOFs, HPAs, adsorption, photocatalytic degradation, catalytic reduction

INTRODUCTION

The increasing contamination of freshwater resources poses a serious global challenge, with significant risks to human health, ecosystems, and socio-economic development. Rapid industrialization, urbanization, agricultural activities, and climate-related disturbances have intensified the discharge of hazardous substances into water bodies, resulting in widespread water pollution.[1,2] Exposure to contaminated water is associated with severe health risks, including waterborne diseases, organ toxicity, carcinogenic effects, and long-term ecological degradation.[3] Water pollutants can be broadly classified into inorganic contaminants (such as heavy metal ions), organic pollutants (including dyes, pharmaceuticals, pesticides, and industrial chemicals), and biological contaminants.[4,5]

Many of these pollutants are persistent, toxic, and resistant to conventional treatment processes, making their effective removal particularly challenging. Organic pollutants are of special concern due to their mutagenic and carcinogenic properties, even at low concentrations, while non-degradable heavy metals tend to accumulate in living organisms and the environment.[7] Conventional water treatment techniques (Fig. 1), encompassing processes such as coagulation, sedimentation, filtration, and disinfection via chlorination, frequently prove inadequate for the complete abatement of contemporary contaminants, particularly those existing at trace concentration levels.[4,8]

Consequently, advanced remediation methodologies have garnered substantial focus due to their enhanced efficacy in eliminating persistent pollutants; these methods include sorption, membrane technologies, advanced oxidation processes (AOPs), and biological degradation.[9] Among these processes, adsorption is distinguished by its operational simplicity, economic viability, and broad applicability across diverse pollutant chemistries.[10] The continual evolution of novel sorbent materials, such as high-surface-area activated carbons, crystalline zeolites, highly porous metal-organic frameworks (MOFs), and

sustainable biopolymers, represents a critical research thrust aimed at optimizing contaminant removal efficiency.[11–13]

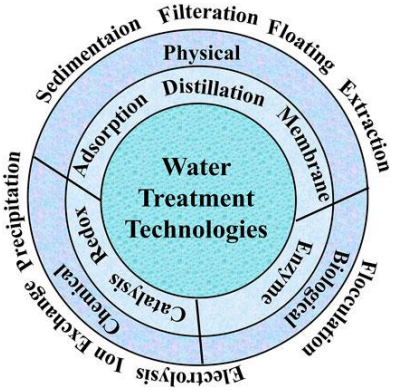

Figure 1: Common water treatment technologies[6]

Cellulose, a naturally abundant, renewable, and biodegradable polymer, has emerged as an attractive platform for the development of sustainable water treatment materials. It is a linear polysaccharide composed of β-D-glucose units linked via β-(1→4) glycosidic bonds and can be sourced from plants, tunicates, microorganisms, and other lignocellulosic biomass (Fig. 2).[14] Its high surface area, mechanical strength, and reactive hydroxyl groups facilitate chemical functionalization and interaction with a variety of pollutants.[15,16] These properties make cellulose an ideal precursor for a range of advanced materials tailored for water purification.[17,18]

Cellulose-derived functional composites for water remediation include several highly efficient types, such as tailored cellulose beads, cellulose-based metal-organic frameworks (CelloMOFs), and heteropolyacid (HPA)-based hybrid composites. Cellulose beads are typically produced as spherical or macroporous microparticles, obtained through the regeneration or chemical modification of native cellulose. Their high specific surface area, mesoporous structure, and facile surface functionalization make them highly effective for the adsorption of metallic ions, anionic dyes, and persistent organic pollutants.[19,20]

CelloMOFs are advanced hybrid nanocomposites in which the structural versatility, ultrahigh surface area, and tunable pore chemistry of crystalline MOFs are synergistically integrated with the biocompatibility and mechanical strength of the cellulose matrix. This combination enables highly selective and kinetically favorable capture of target contaminants. Heteropolyacids, such as phosphotungstic acid immobilized on cellulose, exploit the unique polyoxometalate structure, providing strong Brønsted acidity and reversible multielectron redox potential. These features allow efficient heterogeneous catalysis for the oxidative degradation of organic pollutants, while simultaneously enhancing adsorption capacity.[6]

Heteropolyacids (HPAs) are hybrid materials that combine the features of inorganic polyoxometalates with organic components. They have good stability, selectivity, and adsorption capability. HPAs are especially effective at removing heavy metals, organic pollutants, and even some radioactive compounds from water. HPAs are also known for their strong acidity and redox properties and exhibit remarkable catalytic activity in water treatment processes.[21–23] The intrinsic versatility, renewable nature, and chemically tunable structure of cellulose render it an attractive precursor for the development of next-generation water treatment composites. Contemporary research on cellulose-derived composites is increasingly focused on enhancing adsorption capacity, pollutant selectivity, reusability, and environmental compatibility, thereby overcoming the inherent limitations of conventional water purification methods.[24,25]

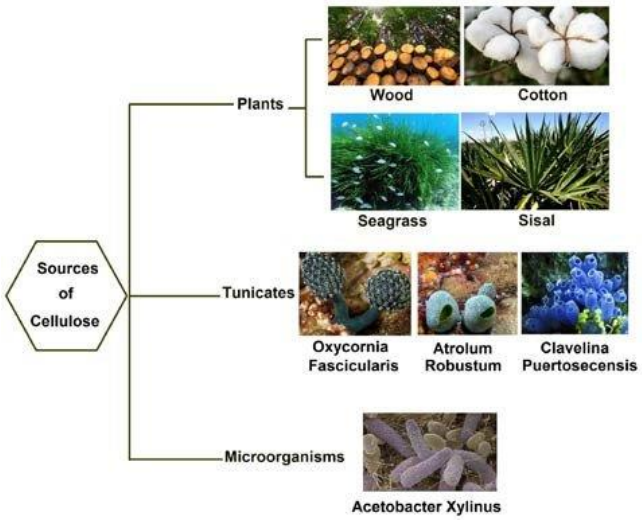

Figure 2: Different sources of cellulose[14]

PROPERTIES AND PREPARATION METHODS OF CELLULOSE BEADS, MOFS AND HPA

Properties and preparation methods of cellulose beads

Cellulose beads are porous, spherical composite materials distinguished by uniform particle geometry, adjustable size, and well-developed internal pore networks, which make them suitable for a wide range of applications, including adsorption, separation, chromatographic packing, catalyst immobilization, and controlled-release systems.[26] Porous beads can be fabricated from both inorganic and organic precursors, such as silica, carbon, synthetic polymers, and natural biopolymers; however, cellulose-based beads are particularly attractive owing to the abundance, renewability, biodegradability, non-toxicity, low cost, and chemical versatility of cellulose.[27,28]

Structurally, cellulose beads exhibit an interconnected meso- and macroporous structure with high specific surface areas, facilitating rapid mass transfer and efficient accessibility of active sites.[26,29] Their spherical morphology improves hydraulic performance, reduces pressure drop, and enables efficient packing in fixed-bed and continuous-flow water treatment systems. Chemically, cellulose beads possess a high concentration of surface hydroxyl ($OH^{\bullet}$) groups, making them hydrophilic and excellent platforms for modification. These reactive sites allow the introduction of diverse charged or chelating functional groups (such as carboxyl, amino, sulfonic, or phosphate).[30,31]

This targeted functionalization is key to boosting their ability to selectively remove pollutants such as heavy metals, dyes, and complex organic molecules through mechanisms including chelation, ion exchange, and various physical interactions.[6] Furthermore, the beads' inherent stability in water and regenerability via mild treatments ensure their successful reuse across multiple cycles, promoting a sustainable water purification process. Despite the functional benefits of cellulose beads, achieving mechanical robustness during fabrication is challenging due to cellulose's inherent insolubility, which stems from extensive intra- and intermolecular hydrogen bonding. Traditional approaches to dissolution, such as the viscose process or the use of complexing agents, often rely on toxic or expensive chemicals, which hinder their large-scale implementation.[26,27]

The earliest description of spherical cellulose bead production was reported in 1951 by Gericke, when a viscous cellulose solution was manually dropped into an aqueous coagulation bath. Since then, numerous methods have been developed to produce beads ranging from 10 μm to 1–3 mm in diameter, using a variety of solvents, regeneration techniques, and pre- or post-treatment steps to tailor their structural and functional properties. More recently, a water-based, one-step method has been proposed for synthesizing hollow spherical cellulose beads, which have potential applications in protein immobilization, water treatment, and drug delivery. The general preparation of spherical cellulose beads

involves three main stages: dissolving cellulose or its derivatives in a suitable solvent, shaping the solution into spherical droplets, and solidifying the droplets into beads through a sol-gel transition.[30,32]

The most promising solution is the low-temperature NaOH-urea aqueous system. This eco-friendly solvent efficiently disrupts the complex hydrogen bond network, allowing for high dissolution efficiency at a low energy cost. Its successful application in preparing various regenerated cellulose forms validates it as a scalable route for producing structurally stable and mechanically improved high-performance cellulose beads.[33–35] The utility of cellulose beads extends to advanced wastewater treatment by serving as robust support matrices for immobilizing active agents, including metal oxides, enzymes, and heteropolyacids.

The integration of cellulose into these beads results in synergistic, multifunctional systems where organic pollutants are first adsorbed and then catalytically or oxidatively degraded. Because cellulose beads combine desirable features such as tailored porosity, adjustable surface chemistry, mechanical stability, ease of regeneration, and environmental sustainability, they stand out as exceptionally promising composites for sustainable water purification applications.[27,36,37]

Forming spherical particles

There are several ways to prepare cellulose beads, but the most commonly used methods include dropping or dispersion processes to shape the beads from a polysaccharide solution. The processes are shown in Figure 3.[30] Dispersion methods do not require complex equipment and enable continuous bead production, whereas dropping techniques necessitate accurate cellulose solution preparation and stringent speed control over the stirring process. The technological factors and a rough macroscopic separation by size make the dropping or dispersion methods of cellulose bead separation practical. For beads larger than 250-500 μm, dropping procedures are utilized to shape them; for beads smaller than this, dispersion methods are employed.[46,47]

In the dropping method, the process of producing spherical droplets of a polysaccharide solution and solidifying them in a nonsolvent coagulation bath can yield beads. However, when a cellulose or cellulose derivative solution is dispersed at a high rotational speed in an immiscible solvent with the opposite polarity, emulsions are formed that can be stabilized with the help of surfactants. The dissolved polysaccharides are present in these emulsions as droplet particles, which can solidify into identical-sized beads.[48]

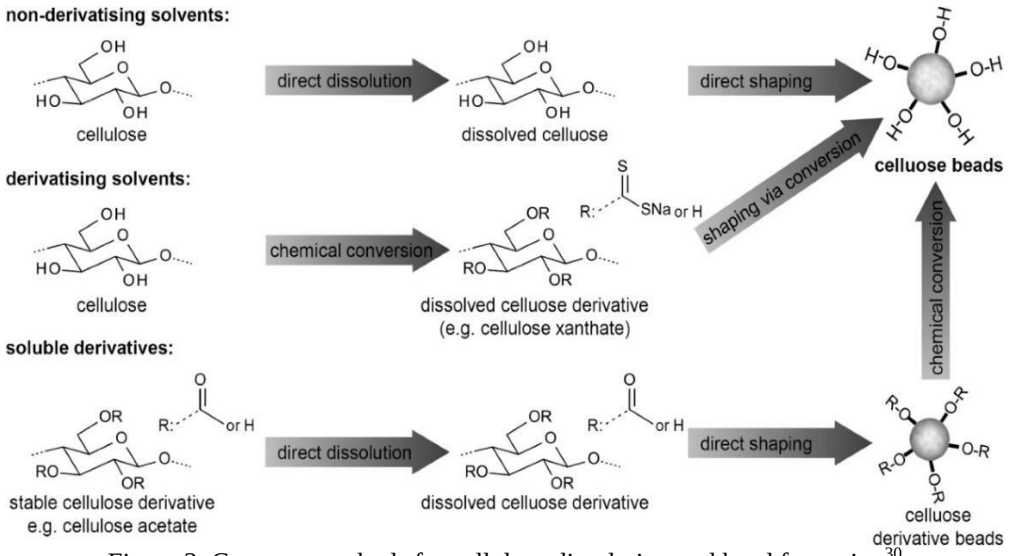

Figure 3: Common methods for cellulose dissolution and bead formation[30]

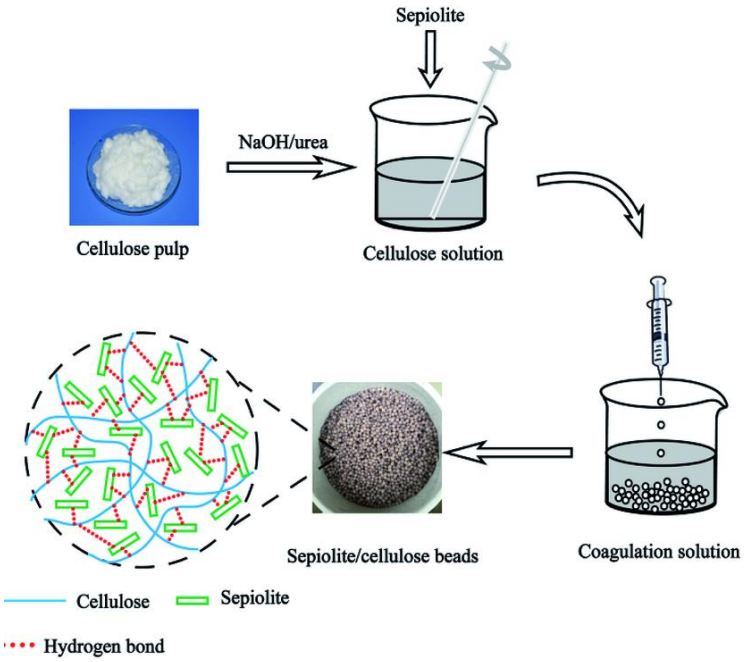

Figure 4: Schematic illustration of the preparation method of sepiolite/cellulose hybrid beads [55]

Dissolution and regeneration of cellulose

Cellulose is characterized by a highly robust network of intra- and intermolecular hydrogen bonds, which renders it insoluble in water and most common organic solvents.[49] Over the years, numerous solvent systems have been developed to dissolve cellulose, enabling its processing into shaped materials such as fibers, films, or beads. These systems can be broadly classified into three categories. Non-derivatizing solvents disrupt cellulose primarily through physical interactions without chemically modifying its hydroxyl groups, and regeneration is achieved by introducing a nonsolvent, such as water, or by altering conditions to induce coagulation, allowing the polysaccharide chains to reassemble into solid structures like beads.[33,50]

Derivatizing solvents temporarily convert cellulose into chemical derivatives that are metastable under dissolution conditions, and regeneration is accomplished by cleaving these intermediates, typically through water addition, pH adjustment, or temperature change, restoring the cellulose in the desired shape. A third approach utilizes stable cellulose derivatives, which are soluble in common organic solvents and often commercially available; these solutions are solidified via coagulation or solvent evaporation in the presence of a nonsolvent. Unlike derivatizing solvents, the substituents of these derivatives remain intact, and an additional step may be required to convert the material into pure cellulose beads. By carefully selecting the dissolution and regeneration route, it is possible to control bead size, morphology, porosity, and mechanical stability, providing a solid foundation for advanced water treatment applications.[32,51,52]

Numerous solvents, including NMMO/H_2O, 68% $ZnCl_2$ solution, onium hydroxides, N,N-dimethylacetamide (DMA) containing LiCl, and ionic liquids like [C4mim] Cl, can be used to dissolve cellulose (Fig. 4). To produce the appropriate bead structure, the dissolved cellulose is usually precipitated in an antisolvent or coagulation bath during the coagulation process.[53,54] Figure 4 displays a generalized synthesis process of sepiolite/cellulose hybrid beads. An identical method was used to create the pure cellulose beads devoid of sepiolite.[55]

The main preparation methods and key properties of cellulose beads relevant to water treatment applications are summarized in Table 1.

Table 1
Preparation methods and key properties of cellulose beads relevant to water treatment applications

Materials	Preparation method	Characteristic properties	Relevance to water treatment	Refs.
Regenerated cellulose beads	Viscose or cellulose carbamate process	Spherical morphology, moderate porosity	Adsorption of dyes and metal ions	30,38
Regenerated cellulose beads	Low-temperature NaOH–urea dissolution and regeneration	High surface area, interconnected meso-/macropores, improved mechanical strength	Enhanced mass transfer, high adsorption capacity, suitability for fixed-bed systems	33,39,40
Functionalized cellulose beads	Surface modification (carboxylation, amination, sulfonation, phosphorylation)	Tunable surface charge, selective binding sites	Selective removal of heavy metals and charged organic pollutants	30,41
Crosslinked cellulose beads	Chemical or physical crosslinking (*e.g.*, epichlorohydrin, citric acid)	Improved mechanical stability, reduced swelling	Reusability and stability during multiple adsorption–desorption cycles	42,43
Composite cellulose beads	Immobilization of HPAs, MOFs, metal nanoparticles, or metal oxides	Combined adsorption and catalytic activity	Adsorption and oxidative degradation of refractory organic pollutants	36,44,45

Metal-organic frameworks

A metal-organic framework (MOF) is a type of crystalline material made up of metal ions or clusters linked to organic ligands. These ligands, which can form potent associations with metal ions, are frequently organic molecules with several binding sites. Due to their high porosity and surface area, MOFs can be precisely modified in terms of their size, shape, and functionality through molecular customization of their pores and channels. Gas storage, catalysis, carbon capture, drug delivery, energy conversion, and water purification are just a few of the numerous applications of MOFs.[56–58]

Figure 5 (A) depicts the basic structure of MOFs with organic linkers and metal ions, while Figure 5 (B) shows other MOFs with their respective metallic clusters and organic linkers. Figure 5 (C) depicts the most common MOF structures. MOFs are generated by the assembly of two components: cluster or metal ion nodes, known as secondary building units, and organic linkers between SBU. This assembly often results in the formation of crystalline structures with significant porous texture development. MOFs have a large pore surface area, micro- and mesopores, and can be customized in terms of pore shape, size, and surface functionality.[59–61]

These features are particularly promising for addressing a variety of difficulties, including the remediation of pollutants through adsorption and catalysis. MOFs have shown great potential for wastewater treatment technologies due to their unique properties, such as distinct pore structures, high flexibility, and excellent selectivity. They have been reported as materials that have high water stability, which are essential for water treatment applications. Due to their high porosity and special adsorbate/adsorbent interactions, MOFs are attractive materials for future applications in pollutant removal from wastewater through adsorption.[62,63] Since MOFs are primarily produced using solvothermal methods, they are excellent choices for eliminating various contaminants from wastewater, including heavy metals, dyes, and antibacterial agents.

Another method for treating wastewater is photodegradation, which does not require additional treatment and promotes complete pollutant removal rather than just a phase transition.[64,65] Because MOFs contain organic linkers, they have a broad absorption spectrum and can generate a charge-separated state that decays quickly, enabling their use in photocatalytic applications.[66,67] Fe-MOFs have been extensively researched for their potential applications in removing air and water pollution, as well as for photodegradation. By interacting with acid sites, forming complexes with pollutants, and facilitating redox reactions, they primarily aid in the elimination of contaminants.

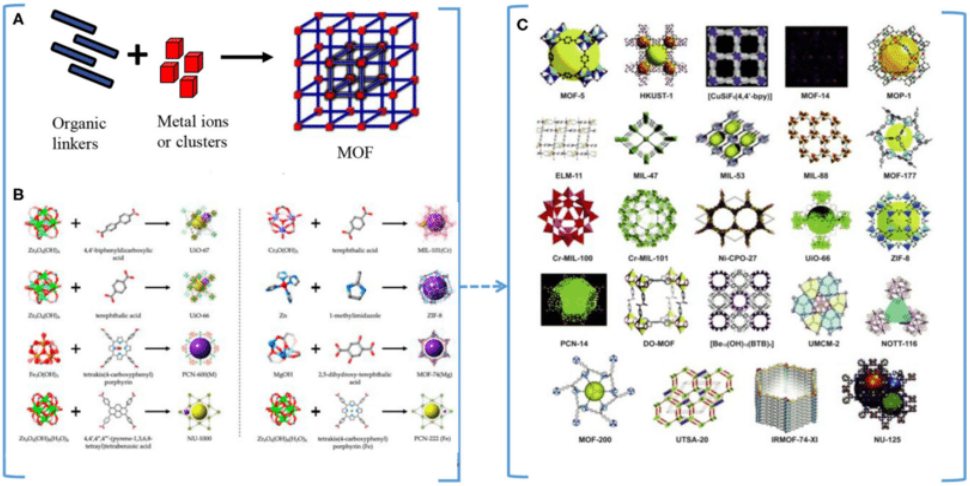

Figure: 5 (A) Basic structure of metallic organic framework (MOF); (B) Examples of different MOF structures with their corresponding metallic clusters and organic linkers; (C) Examples of common MOF structures[61]

Fe-MOFs have been applied for the photocatalytic removal of several water contaminants, including hexavalent chromium, as well as organic pollutants like dyes and antibiotics.[68] MOFs have been utilized in the development of effective MOF-based adsorbents for the removal of organic pollutants from wastewater, in addition to adsorption and photodegradation. Zr/Fe-MOFs/GO (graphene oxide) composites have been developed to efficiently eliminate Orange II and tetracycline hydrochloride at different concentrations. The results indicated that the Zr/Fe-MOFs/GO exhibited respectable robustness and high rates of pollutant removal, also suggesting potential for reusability.[66]

MOFs are not well-suited for large-scale commercialization because of their mechanical instability, fragility, and difficult workability. Researchers have combined MOF particles with other flexible and processable materials, such as polymers, graphene, and mesoporous silica, to create hybrid materials that can overcome these restrictions. However, a major obstacle to the practical implementation of MOFs is still finding substrate materials with sufficient mechanical strength, acceptable processability, low cost, and long-term stability.[69–71]

MOF hybrid materials have demonstrated potential for application in gas separation, energy storage, catalysis, and nanomedicine, among other fields. These hybrid materials are produced by combining existing MOFs with different substances or small molecules using a variety of methods, including covalent alterations, noncovalent interactions, and the use of MOFs as precursors or sacrificial templates.[72] Due to their low cost, biocompatibility, nontoxicity, and abundance of surface groups that are advantageous for MOFs grafting, cellulose and its derivatives are ideal substrate materials for MOFs grafting. When it comes to potential industrial manufacturing and practical application value, they outperform various synthetic polymers, including graphene.

Numerous investigations have documented the process of creating graft-copolymers from cellulose and its derivatives by the application of atom transfer radical polymerization (ATRP) in conditions of homogeneous reaction.[73,74] Numerous other investigations have documented the processes of homogeneous and heterogeneous ring-opening polymerization (ROP) grafting from cellulose and cellulose derivatives, UV grafting onto cellulose and cellulose derivatives, and CuAAC click chemistry grafting of cellulose and cellulose derivatives. These investigations show the promise of cellulose and its derivatives for enhanced cellulose-based materials and gels, as well as for MOF grafting.[75,76] Table 2 shows different types of CelloMOF hybrids with their method of preparation and water treatment application.

Table 2
Types of CelloMOF hybrids with their method of preparation and water treatment application

CelloMoFs	Synthesis method	Water treatment application	References
(i) UiO-type CelloMOFs (*e.g.* UiO-66 and UiO-67)	(a)Solvothermal (b) Hydrothermal	(a) Photocatalytic degradation	[71]
(ii) MIL-type CelloMOFs (*e.g*., MIL-53 and MIL-101)	(a)Solvothermal (b) Microwave	(a) Adsorption (b) Oxidation	[77]
(iii) MOF-5-type CelloMOFs (*e.g*. MOF-5 and MOF-177)	(a) Solvothermal (b) Microwave	(a) Adsorption (b) Catalytic degradation	[78,79]

Preparation of cellulose-MOF composites

Cellulose-metal organic framework composites are made by dispersing MOFs in the supported phase using cellulose or its derivatives as the substrate.[77] A variety of functional composites can be produced by the *in-situ* growth or *ex-situ* grafting of MOFs made possible by the fibrous cellulose matrix. Figure 6 displays the synthesis of Cello-MOFs by utilizing the *in-situ* and *ex-situ* methods. The cellulose-based materials were treated with the addition of MOF precursors (metals or linkers) in an *in-situ* synthesis approach. Before processing, MOF precursors can be added to cellulose materials or cellulose forms like foams and aerogels. The resulting cellulose-MOFs can then be used to create foams and aerogels by traditional techniques, including freeze-drying and casting (Fig. 6).

In the second *in-situ* method, a cellulose scaffold was combined with the MOFs precursors (metal ions or organic linkers) (Fig. 6). After the use of conventional synthesis techniques like solvothermal or hydrothermal methods, MOFs grow *in situ* into the fibers. The best application for MOF precursors is unprocessed cellulose fibers. This is because most MOFs that were made need organic or high-temperature solvents. Certain circumstances might damage cellulose forms. Additionally, the cellulose-MOF fibers can subsequently be processed conventionally to take on the appropriate shape (Fig. 7).[71]

It is possible to alter cellulose fiber to ensure a high level of metal-fiber contact. The reactive dye method (RDM), polydopamine, cyanuric chloride-modified thiol reaction, and esterification can all be used to accomplish the modification.[80] A few advantages of the *in-situ* synthesis method include its straightforward synthesis process, fast growth of MOFs, and robust interactions with cellulose fibers. When MOF precursors crystallize, cellulose helps to control the morphology, lower the size of the crystal particles, and accelerate the formation of the crystals. The mechanical properties of cellulosic forms can also be enhanced by these methods.[81,82]

The distribution of MOFs can be homogeneous; however, controlling the MOFs' loading into cellulose form using the *in-situ* method is challenging. The products of MOFs in cellulose materials are typically low; the loading can be increased by adding MOF precursors one after the other or by using techniques like layer-by-layer (Fig. 7). The addition of metals or organic linkers may cause changes in the conformational structure of cellulose. Synthesized MOFs can be deeply embedded into cellulose fibers, preventing the diffusion of reactants or adsorbates into MOF crystals.[71,77]

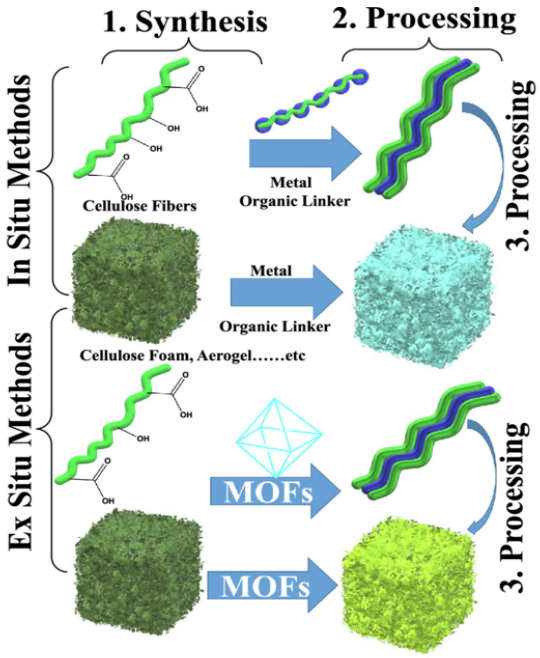

Figure 6: Synthesis of Cello-MOFs using *in-situ* and *ex-situ* methods[71,77]

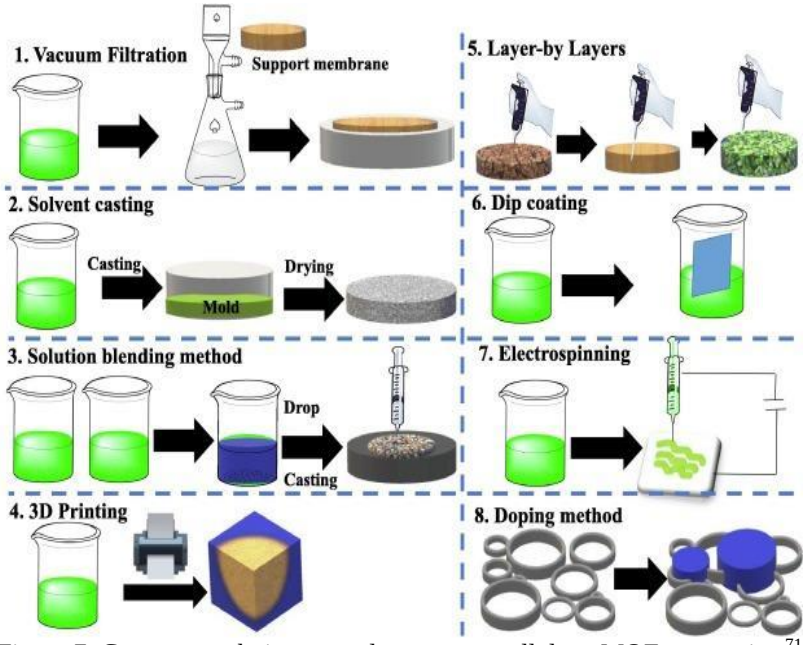

Figure 7: Common techniques used to prepare cellulose-MOF composites[71]

The *ex-situ* approach is the second strategy for producing CelloMOFs (Fig. 6). Using *ex-situ* techniques, synthesized MOFs are loaded into cellulose fibers or cellulose form (Fig. 6). These techniques require several steps to be taken. The synthesis of MOFs using traditional techniques is the first stage. The synthetic MOFs are added to cellulose fibers or cellulose form in the second stage. The final phase involves utilizing standard processing techniques to transform cellulose-MOFs into the required form (Fig. 7).[71]

Another possible option is using MOF solutions to dope cellulose-structured supports. One benefit of using the *ex-situ* approach is that it allows for the control of MOF loading into cellulose.[77,83,84] In

comparison with *in-situ* synthesis methods, it allows high loadings (Fig. 6). Since most MOF crystals are found on the cellulose form's surface, reactants and chemical reagents can easily reach them. As a result, the materials synthesized had a high adsorption capacity and a large surface area. However, these techniques required several processes that took time and chemical reagents. The method's shortcomings include limited uniform diffusion within the cellulose network and severe MOF agglomeration.[85,86]

Heteropolyacids

Heteropolyacids (HPAs) are a family of inorganic compounds made up of oxygen atoms joined in a polymeric structure and metal cations, typically transition metals (Fig. 8). They have several uses in diverse industries and are renowned for their special qualities. Acidic hydrogen-oxygen-addenda atoms like silicon and heteroatoms like vanadium (V), molybdenum (Mo), and tungsten (W) make up HPAs. These compounds, which have three or more transition metal oxyanions connected by common oxygen atoms, are a subclass of polyoxometalates. Keggin-type HPAs, which have the anions of $[XM_{12}O_{40}]^{n-}$, where X is a heteroatom (such as P or Si), and M is a metal ion (such as W^{6+} or Mo^{6+}), are the most prominent type of HPAs (Fig. 8). Phosphotungstic acid ($H_3PW_{12}O_{40}$) is the most predominant Keggin-type HPA among them.[23,87]

Due to its high solubility in water and ability to effectively dissociate protons, this specific HPA makes it easier to interact with a variety of substrates, thus speeding up reaction rates. Keggin-type HPAs possess unique qualities, such as strong acidity, low charge density, and good symmetry, making them useful as both homogeneous and heterogeneous catalysts. Their reusability in heterogeneous systems is improved by their ease of separation from the reaction mixture. Other examples of heteropolyacids include silicotungstic acid ($H_4SiW_{12}O_{40}$), phosphomolybdic acid ($H_3PMo_{12}O_{40}$), and silicomolybdic acid ($H_4SiMo_{12}O_{40}$).[88]

Due to their unique redox characteristics and strong Bronsted acidity, HPAs are a type of eco-catalysts that have been extensively used in a variety of homogeneous and heterogeneous processes. Two main factors contribute to the strong Bronsted acidity of HPAs: the negative charge is dispersed over a large number of atoms in the polyanion, and the double-bond character of the M-O_d bond polarizes the negative charge of O_d to M, resulting in a less distributed negative charge over the polyanion's outer surface.[87] For HPA-catalyzed processes, the standard mechanism of Bronsted acid catalysis, which includes two catalysis types: bulk and surface, is well known.

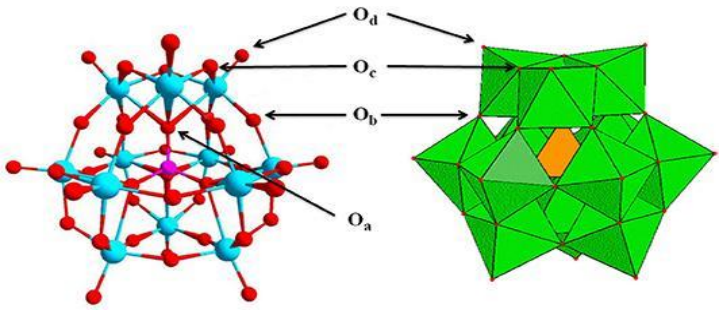

Figure 8: The common structure of Keggin-type HPA[87]

HPAs have been employed as catalysts in numerous chemical reactions, such as heterocycle synthesis and hydrolytic depolymerization of cellulosic biomass.[21] The superior catalytic activity of HPAs is well-known, ranging from three to one hundred times that of regular organic and inorganic acids. The hydrophobic solid catalysts HPAs are also effective at withstanding water in biphasic reaction systems that include an aqueous phase. Through various processes, including adsorption, photocatalytic degradation, and catalytic reduction, they can assist in the treatment of water. Adsorbents, or high-performance ammoniac acids, can draw in and bind to impurities in water. During this process, pollutants are chemically or physically bonded to the surface of the HPA particles. Adsorption enables HPAs to

successfully remove organic molecules, heavy metals, and other contaminants from water, thereby improving its quality.[22,87]

HPAs can initiate chemical processes when they encounter light, due to their photocatalytic properties. They can break down organic pollutants in water by acting as photocatalysts. Reactive oxygen species (ROS), which are produced when HPAs are exposed to light, can break down organic molecules into less dangerous and simpler compounds. Moreover, HPAs can catalyze reduction reactions, which makes it possible to change poisonous heavy metals like mercury or chromium into less harmful forms that are simpler to remove from water.[87,89]

A photocatalyst absorbs light energy and produces several kinds of free radicals, which are highly reactive chemical entities. However, the fact that photocatalytic processes can only use less than 5% of solar light limits the practical application of HPAs. Current research is exploring the potential of HPAs as photocatalysts for the degradation of organic pollutants in water treatment.[90] To show the potential of heterogeneous photocatalysts for environmental applications, different photocatalytic oxidation and reduction processes of HPAs in water are compared. Furthermore, although it necessitates high calcination temperatures, the production of light-activated hydroxyapatite (HAP) photocatalysts is a somewhat straightforward process that can be applied to the treatment of water, providing benefits like low cost.[91,92]

When HPAs and metal-organic frameworks (MOFs) are combined, they create materials with improved stability, acidity, and catalytic activity. This combination shows promise for a variety of catalytic reactions, such as acid-catalyzed reactions and oxidation, by allowing the synthesis of materials with specific characteristics. The porous nature of MOFs allows HPAs to disperse over a vast surface area, increasing catalytic efficiency. Furthermore, MOFs' stability reduces deactivation and leaching into treated water by keeping HPAs within the material. HPAs become more stable and reusable when they are immobilized in MOFs, which lessens the requirement for frequent replacement. Ongoing research and development on HPA-MOF-based materials is driven by their potential in a variety of catalytic applications.[93–95]

Preparation of heteropolyacids

Since heteropolyacids have strong oxidizing capabilities and can remove a wide range of contaminants, they are widely used in water treatment applications. Usually, they are prepared by dissolving a metal salt precursor in water or an appropriate solvent, such as tungsten, molybdenum, or vanadium salts. To guarantee complete dissolution, the solution is subsequently treated under carefully monitored circumstances with a strong acid, such as sulfuric acid. Through a series of chemical reactions, the addition of a strong acid produces heteropolyacids. After the strong acid is added, the heteropolyacids may precipitate out of the solution. The use of filtration or centrifugation is required to separate the precipitate from the liquid phase.[96,97] Then, the precipitate is cleaned of any contaminants or leftover reactants using an appropriate solvent, like water or alcohol. To produce the finished heteropolyacids product, the cleaned precipitate is dried under carefully controlled conditions, such as low temperature or vacuum. Heteropolyacids are easily isolated from the products in both homogeneous and heterogeneous systems by a straightforward extraction procedure. They have shown promise in catalyzing a variety of reactions, including hydration and dehydration, which are crucial for the treatment of water.[98]

Llewelyn suggested that the preparation of heteropolyacids involves the polymerization of octahedrally coupled metal oxides around a heteroatom in an acidic solution. The exterior oxygens, which are oriented toward the W or Mo atoms, inhibit the heteropolyacid from building bigger structures. The resulting MO_6 octahedra are somewhat deformed, allowing the heteroatom to be accommodated.[98,99] External oxygens are weakly basic and can only form weak bonds with protons. Keggin HPAs are easily produced at ambient temperature. The easiest technique to produce a Keggin-type heteropolyacid (HPA) is by acidifying phosphoric acid (H_3PO_4) and sodium tungstate (Na_2WO_4), by the following reaction:

$$12W04^{-2} + HP04^{-2} + 23H^{+} \text{ —►} PW_{12}O_{40}^{-3} + 12H_2O \quad (1)$$

Since the HPA isomer is only stable at a pH of 1, the solution needs to be lower than 1. Moreover, it can be simply separated via solvent extraction to produce a white powder. However, there is a limited number of methods that have been reported in the literature about the preparation of HPAs, using different solvents and catalysts.

ADSORPTION CAPABILITIES OF CELLULOSE BEADS, CelloMOFs AND HETEROPOLYACIDS

Adsorption is considered a simple, low-cost, non-destructive method that effectively removes a wide range of contaminants from wastewater and water. The most popular method for eliminating organic materials, heavy metals, and other contaminants is to use activated carbon. Despite its great efficiency, activated carbon in wastewater treatment is restricted because of its high cost and intricate regeneration procedures. Still, adsorption can be a feasible and easy method for treating wastewater due to the discovery of several inexpensive adsorbents with excellent efficiency for the removal of dyes and heavy metals from industrial effluents. The choice of adsorbents is influenced by several variables, such as renewability, availability, cost-effectiveness, capacity, and efficiency.[11,100–102] Cellulose beads, CelloMOFs, and heteropolyacids are some of the materials that have been used as adsorbents in water treatment applications.[6]

Cellulose beads

Cellulose beads possess excellent adsorptive capabilities due to their unique structure and properties. The key factors contributing to their outstanding adsorption capabilities are described below.[28,103,104]

Porous structure

Cellulose beads are widely recognized as effective and environmentally friendly adsorbents due to their highly porous structure and large surface area.[105,106] Their specific surface area generally falls between 100 and 500 m^2/g, although specially cross-linked forms can reach values as high as 900 m^2/g.[106,107] The high porosity of these beads, often above 90%, allows water and pollutants to move easily through the material, while the well-developed pore structure provides many accessible sites for adsorption.[108,109] As a result, cellulose beads show strong removal performance, with reported adsorption capacities of up to 570 mg/g for heavy metals and more than 1,500 mg/g for organic dyes.[110] Additionally, the numerous hydroxyl groups on the cellulose surface enable chemical modification, such as introducing amino or carboxyl groups, which can further enhance adsorption efficiency and selectivity toward specific contaminants.[108,111]

Hydrophilic nature

Cellulose beads leverage the inherent properties of cellulose, such as its hydrophilicity, to effectively remove water-soluble contaminants. The material's strong affinity for water molecules makes it an efficient adsorbent for a variety of polar substances and pollutants found in water, including organic compounds, dyes, and heavy metals. These characteristics enable cellulose beads to effectively bind and remove impurities from aqueous solutions.[112]

Functionalization potential

Cellulose beads can be easily modified to improve their adsorption properties and allow for the selective adsorption of specific compounds due to their functionalization capabilities. This increases the overall efficiency of the process in the treatment of water. Cellulose and its derivatives can be used to make functional cellulose beads, which have many uses, such as medication delivery and the adsorption of heavy metals and other pollutants.[113]

Adsorption mechanism

Studies have shown that cellulose beads display excellent adsorption efficiency in two ways: chemical adsorption, which involves chemical reactions between the adsorbent and the adsorbate, and physical

adsorption, which involves contaminants being drawn to the surface of the beads by weak van der Waals forces.[108,114] For example, literature has demonstrated that cellulose/chitin beads use both chemical and physical adsorption mechanisms to successfully adsorb heavy metal ions, including Pb^{2+}, Cd^{2+}, and Cu^{2+}.[27] The good recoverability and adsorption efficiency of the functionalized cellulose beads have also been observed, demonstrating the potency of the adsorption mechanisms used by the modified cellulose beads. Thus, acting by two different adsorption methods enables cellulose beads to effectively capture a wide range of pollutants.[115]

Ion exchange capacity

Ions can be extracted from water by cellulose beads by adsorption and ion exchange techniques. There are several techniques to determine the ion exchange capacity of cellulose beads, such as porosity-dependent tests and acid-base titration. Peptide and other biomolecule chromatography on a laboratory or industrial scale can benefit from the superior flow characteristics, mechanical stability, and chemical resistance of Cellofine ion exchangers, which are based on spherical particles made from crosslinked cellulose. Based on the type of sorbent, cellulose beads have an ion exchange capacity ranging from 0.13 to 0.63 mmol/mL. To ascertain whether bead cellulose derivatives are appropriate for a given application, measurements of their pK values and ion exchange capacity can be made.[116–118]

Regeneration process

Repeated use of cellulose beads is made possible by their regeneration, which lowers the amount of wastes produced. Studies have revealed that, even after numerous adsorption-desorption cycles, cellulose beads with a high carboxyl group concentration retained a high adsorption capacity, indicating their durable performance. Furthermore, it is possible to remove impurities from cellulose beads using eluents such as NaOH, suggesting that the beads may be regenerated and be reused. Cellulose beads are a viable solution for lowering waste production and enhancing environmental sustainability due to their effective regeneration and high adsorption capacity.[119]

Cellulose-metal organic frameworks

For water treatment applications, cellulose–metal-organic framework hybrids (celloMOFs) have the advantage of being porous materials that combine the benefits of metal-organic frameworks (MOFs) and cellulose. These materials are promising for a variety of water treatment procedures due to their special adsorptive capabilities. Below is a detailed clarification of the characteristics that support the adsorption capabilities of celloMOFs.[71,77,120]

High surface area

Integrating metal-organic frameworks (MOFs) into cellulose materials significantly improves the surface properties of the resulting CelloMOFs. While pristine cellulose typically shows very low BET surface areas (often below 10 m^2/g), MOF incorporation transforms these materials into highly effective adsorbents. For example, ZIF-8/cellulose nanofibril composites have achieved surface areas as high as 1014 m^2/g.[121] In ZIF-67/triacetyl cellulose aerogels, increasing the MOF content from 18 to 51 wt% raised the surface area from 560.20 to 734.60 m^2/g.[122] Similar enhancements have been reported for HKUST-1/pulp fiber (314 m^2/g),[123] UiO-66/bacterial cellulose (267 m^2/g)[124] and MOF-199/carboxylated cellulose composites (264.83 m^2/g).[125] These improvements arise from the high intrinsic porosity of MOFs effectively transferred to the cellulose matrix.

Tunable porosity

The enhanced porosity of celloMOFs is central to their effectiveness as filtration materials, with some composites exhibiting total porosities up to 98.96%.[126] Pristine cellulose typically shows low BET surface areas (below 10–30 m^2/g), but MOF incorporation greatly increases internal free volume; for instance, ZIF-8/cellulose nanofibril composites have reached surface areas of 1,014 m^2/g.[2,6] This property is tunable through MOF loading, as demonstrated in ZIF-67/triacetyl cellulose aerogels, where increasing

MOF content from 18% to 51% raised the surface area from 560.20 to 734.60 m^2/g. Optimized synthesis also significantly increases pore volume, from ~0.41 mL/g in raw cellulose to over 6.0 mL/g in MOF-based composites, resulting in combined micro- and mesoporous structures that support selective adsorption and efficient mass transport.[127–129]

Chemical functionality

The chemical activity and selectivity of celloMOFs are largely controlled by the choice of MOF linkers and the introduction of functional groups into the framework.[130] Functionalities, such as hydroxyl, amino, sulfonic, and carboxyl groups, provide targeted interaction sites that improve adsorption toward specific water contaminants.[131,132] For example, amino-functionalized systems like UiO-66-NH_2/cellulose show strong affinity for anionic dyes and heavy metals due to enhanced electrostatic interactions and hydrogen bonding.[133] These modifications not only increase adsorption efficiency, but also enable the removal of contaminants that are otherwise difficult to treat, highlighting the synergistic effect between the MOF structure and chemically active linkers in aqueous pollutant removal.[134]

Affinity for different contaminants

CelloMOFs show a strong ability to capture a wide variety of contaminants, including microplastics, organic pollutants, dyes, heavy metals, and pharmaceuticals.[135] This strong adsorption performance arises from the combined action of metal ions and organic linkers in the MOF structure, which interact with pollutants through coordination bonding, electrostatic attraction, and hydrogen bonding.[136] As a result, celloMOFs can efficiently remove many emerging contaminants, such as oils, pesticides, phenolic compounds, and both positively and negatively charged dyes. Their well-developed internal structure provides many accessible interaction sites, allowing for high adsorption capacities and often fast removal rates, which makes celloMOFs highly promising materials for advanced water treatment applications.[137]

Regenerability

Cellulose–metal organic framework hybrids (celloMOFs) are particularly attractive for water treatment because they can be easily regenerated and reused. Pollutants adsorbed on these materials can be removed using simple methods, such as solvent washing, changes in pH, or mild heating, allowing the same material to be used over several adsorption–desorption cycles.[135] This reusability makes celloMOFs both cost-effective and environmentally friendly for wastewater treatment applications.[138] In addition, the cellulose support offers extra sustainability benefits, as it can be reprocessed into other useful forms, such as aerogels, textile fibers, or transparent films after use.[139] By combining a renewable cellulose framework with a recyclable MOF component, celloMOFs represent durable and sustainable materials for tackling water pollution challenges.[132]

Heteropolyacids

Heteropolyacids (HPAs) have demonstrated promising adsorptive properties in a range of applications, including catalysis and water treatment. Research results highlight the potential uses for HPAs in water treatment by showcasing their adsorptive and catalytic properties.[140–143]

High surface area

Heteropolyacids (HPAs) have a unique metal–oxygen framework that provides well-defined active sites for capturing different pollutants. On their own, bulk HPAs usually have very low surface areas (about 1–10 m^2/g), and their performance strongly depends on their chemical composition. For instance, phosphotungstic acid (HPW) often shows better adsorption behavior than silicotungstic acid (HSiW) because of differences in the coordination environment within the polyoxometalate structure.[144] To improve their effectiveness, HPAs are commonly supported on porous materials, such as zirconia-modified alumina or clays, which greatly increases surface area and improves access to active sites.[145] These supported systems show enhanced removal of contaminants like heavy metals and organic dyes due to improved mass transport and stability.[146]

Strong acidity

Heteropolyacids (HPAs) are widely recognized for their strong Brønsted acidity, which plays a crucial role in their effective interaction with various pollutants. This strong acidity promotes adsorption through processes such as complex formation, ion exchange, and electrostatic attraction, making HPAs both efficient adsorbents and active catalysts in environmental applications.[147] When HPAs are supported on materials like kaolin, strong interactions between the HPA species and the support have been shown to improve chemical adsorption performance.[148] In addition, combining HPAs with porous supports, such as alumina or organic–inorganic hybrid materials, allows their acidity and surface properties to be fine-tuned, which can further enhance pollutant removal efficiency. Overall, the pronounced acidity of HPAs is central to their effectiveness and underpins their wide use in catalytic and environmental remediation processes.[149]

Selectivity

Heteropolyacids (HPAs) are highly effective for water treatment because their properties can be tailored to target specific pollutants based on size, charge, or chemical nature.[148] By adjusting the HPA framework through the choice of metal ions or heteropoly salts, active sites can be optimized for capturing toxic substances.[150] Their adsorption selectivity is also influenced by solution pH, which affects both the contaminant's ionization and the surface charge of the HPA material.[151] This pH responsiveness enables targeted removal of pollutants, such as heavy metals or organic dyes, by exploiting electrostatic interactions or complexation under controlled conditions.[152]

Regeneration potential

Heteropolyacids (HPAs) are valued for being both cost-effective and environmentally friendly, as they can be repeatedly regenerated and reused in water treatment and catalytic processes.[153] Their adsorptive properties can be restored through simple thermal or chemical treatments, including solvent washing and pH adjustment, allowing them to maintain performance over many cycles. This recyclability lowers remediation costs and reduces secondary waste, while the stability of the structure ensures long-term effectiveness in removing pollutants from water.[152,154]

Stability

The exceptional chemical and thermal resilience of heteropolyacids (HPAs) is fundamentally rooted in the structural robustness of the Keggin and Wells-Dawson frameworks, which allows these materials to maintain their performance across diverse pH ranges and fluctuating temperatures.[23,154] This inherent stability ensures that their adsorptive and catalytic functionalities remain intact even under the corrosive or demanding conditions often encountered in industrial water treatment. Furthermore, HPAs exhibit significant resistance to oxidative degradation and can withstand high thermal loads, making them ideal candidates for use as heterogeneous photocatalysts and long-lasting adsorbents. When immobilized on solid carriers, such as silica or alumina, the leaching of active acidic species is minimized, further extending their operational lifespan and suitability for large-scale environmental remediation.[155]

PHOTOCATALYTIC AND CATALYTIC REMEDIATION ABILITIES OF CelloMOFs AND HETEROPOLYACIDS

Promising approaches for treating water include photocatalytic degradation and catalytic remediation, which are especially effective in eliminating organic contaminants from wastewater. Photocatalysis is a process that uses photocatalysts to break down pollutants when exposed to light. It has become more popular since it works well in moderate environments and is sustainable. However, for practical implementation, issues like low degradation efficiency and reusability must be resolved. Numerous studies have demonstrated how well photocatalysis works to break down and mineralize stubborn pollutants in water, such as hormone disrupting agents, biocides, and synthetic dyes. Furthermore, it has

been suggested that the removal of emergent water contaminants could be improved by using photocatalytic materials in optimized packed-bed photoreactors.[156,157]

The method known as catalytic reduction makes use of a catalyst to help reduce the amounts of contaminants in water. Various materials can be used to create the catalyst, including noble metals, dendritic polymers, and palladium-based catalysts. Various techniques can be used to accomplish the reduction, including hydrogenation and electrocatalysis. Catalysts can be used to increase the reduction process and lower their energy requirements. Although there are still obstacles and knowledge gaps in the development of catalytic reduction techniques, there are also fresh avenues for investigation and innovation in this area. CelloMOFs, and heteropolyacids have been used as catalysts for photocatalytic and catalytic remediation in water treatment applications.[157,158]

CelloMOFs

CelloMOFs have been investigated for their potential use in photocatalytic and catalytic reduction processes. These materials possess a large surface area, which promotes rapid catalytic degradation or high adsorption of pollutants. Research has shown that incorporating cellulose materials into photocatalysts, such as MOFs, improves their photocatalytic capabilities. Because of their adaptability at the compositional, chemical, and porous structural levels, MOFs including CelloMOFs display photocatalytic capabilities that make them effective for a variety of reduction processes, like the reduction of Cr(VI) to Cr(III). These materials are promising for a variety of catalytic applications due to their adaptability, which provides numerous options to adjust light harvesting, charge mobility, and transfer.[159,160]

Photocatalytic degradation abilities of CelloMOFs

It has been demonstrated that incorporating MOFs into cellulosic materials improves photocatalyst efficiency for wastewater treatment and environmental remediation. The specific degrading capacities of CelloMOFs are subject to variation according to the synthesis processes, composition, and structure employed. When exposed to light, CelloMOFs' exceptional photocatalytic qualities enable them to effectively break down a variety of organic contaminants. Photons are absorbed by CelloMOFs in the presence of light, and this excites the material's electrons. The energy from these excited electrons can be transferred to oxygen molecules or other reactive species, producing highly reactive radicals such as hydroxyl radicals (OH). These radicals can efficiently decompose organic substances into smaller, less toxic molecules or mineralize them entirely into carbon dioxide and water. CelloMOFs have been shown to be highly efficient nano heterojunctions for the inactivation of harmful bacteria and photocatalytic destruction of organic pollutants. Additionally, they have been used as filters and catalysts for the reduction, oxidation, and catalytic degradation of organic contaminants. Overall, CelloMOFs may be an inexpensive and extremely effective way to stop the environment from becoming overly contaminated with organic pollutants and harmful germs.[4,71,81,160]

Catalytic reduction abilities of CelloMOFs

CelloMOFs can catalyze reduction processes and convert hazardous compounds into non-toxic ones. By acting as active sites for catalytic reactions, the metal centers in the MOF structure can encourage the reduction of pollutants through various processes, including transfer hydrogenation, hydrogenation, and dehydrogenation. Numerous processes, such as the electrochemical reduction of CO_2, the selective catalytic reduction of NOx, the degradation of OPDs, and the electrocatalytic reduction of carbon dioxide, have all been studied with MOFs as catalysts. When it comes to the removal, separation, or degradation of pollutants, such as heavy metals, CO_2, CH_4 and OPDs, MOFs perform better than traditional materials.

The activity and selectivity of CelloMOFs as catalysts are strongly influenced by the selection of metal nodes and their coordination environment, as well as by the composition of the organic linkers and their interactions with the metal nodes. Furthermore, variables including catalyst loading, co-catalyst presence, and reaction conditions (temperature, pressure, solvent) might affect the total catalytic performance. There are ongoing efforts to improve CelloMOFs' catalytic efficiency in reduction

processes. To enhance electron transfer kinetics and boost the quantity of active sites, techniques include adding dopants or co-catalysts. Changes to the electrical and structural characteristics of CelloMOFs, such as functionalization or the introduction of defects, can also improve catalytic efficacy.[77,161,162]

Heteropolyacids

Photocatalytic degradation abilities of heteropolyacids

Heteropolyacids are capable of photocatalytic degradation because they may absorb light energy and produce reactive species, such as holes (h^+), superoxide radicals ($\bullet O^{2-}$), and hydroxyl radicals (•OH). These reactive species oxidize organic contaminants to start degradation processes. Both visible and ultraviolet (UV) light may be absorbed by HPAs, which excite electrons and form electron-hole pairs that are involved in redox processes. Superoxide radicals can assist in the degradation process through oxidation processes. However, the highly reactive hydroxyl radicals produced can also break down chemical bonds in organic contaminants. Several parameters, such as the composition of metal cations, the structure of polyoxometalate anions, the pH of the reaction media, and the selection of light source and intensity, can affect the photocatalytic degradation capabilities of HPAs.[158,163]

Heterostructured HPAs hold great potential as extremely effective photocatalysts for the treatment of wastewater contaminated with dyes, as research has shown.[96] Additionally, the larger surface area of HPAs may boost their catalytic activity by providing more contact between the catalyst and the substrate. Additionally, as demonstrated by the investigation of the homogeneous photocatalytic destruction of phenol by heteropolyacid salts under artificial UV irradiation, the selection of light source and intensity can affect the photocatalytic performance of HPAs.[96]

Catalytic reduction abilities of heteropolyacids

In water treatment applications, HPAs have exhibited noteworthy catalytic reduction capabilities. These compounds are made up of oxygen atoms encircling metal cations, usually transition metals, in a polyoxometalate structure. The strong redox and acidity characteristics of HPAs make them efficient catalysts for a variety of processes, including the removal of contaminants from water. There are various reasons why HPAs are effective at catalyzing reduction in the water treatment process. First off, because of their high acidity, the pollutant molecules can be protonated, which makes reduction easier. Moreover, HPAs have redox-active metal centers that are capable of reversible redox reactions, which enable them to take part in catalytic reduction processes involving electron transfer. The process by which HPAs catalyze reduction can differ based on the type of pollutant and HPA being utilized.[21]

Occasionally, HPAs take on the role of electron donors, providing electrons directly to pollutant molecules. In other situations, reactive species like free radicals that are produced by HPAs may react with the pollutant to aid in its elimination. The hydrolytic depolymerization of cellulosic biomass and the conversion of biomass into usable chemicals and biofuels have both been made easier with the use of HPAs as heterogeneous catalysts. They can be easily extracted from products in both homogeneous and heterogeneous systems using a simple extraction procedure. HPAs have special qualities that make them useful for a variety of catalytic applications, including the valorization of lignocellulose and the catalysis of organic processes in water. These qualities include superior solubility and great thermal stability.[87]

CONCLUSION

Various techniques have been developed for the preparation of cellulose beads, CelloMOFs, and heteropolyacids. The preparation techniques and materials used depend on the desired properties of these materials. Cellulose beads have shown considerable promise in water treatment applications due to their unique qualities, such as their porous structure, high surface area, and great adsorption capability. These beads can effectively eliminate various contaminants from water sources, including heavy metals, dyes, organic pollutants, and microbes. The porous nature of the beads allows impurities to be trapped inside the bead structure, essentially eliminating them from the water. The incorporation of MOFs has been reported to improve the porosity, mechanical, and thermal properties of cellulose-based materials. The type of MOFs used depends on the intended application, and cellulose-MOFs integrate the benefits of

both materials, allowing them to have more uses than either material alone. MOFs also prevent the formation of aggregation seen with high cellulose concentrations and restrict the interaction of the nanocellulose surface's numerous hydroxyl groups.

Heteropolyacids exhibit strong magnetism, photocatalytic capabilities, conductivity, and redox activity; however, their limited applications are attributed to their dissolution in polar solvents and poor surface area. To improve their performance, numerous modification strategies, including grafting, have been developed. One promising technique is to load heteropolyacids into porous materials with a high surface area, such as MOFs, cellulose-based materials, and metal oxides like SiO_2. However, there are no reports on the incorporation of heteropolyacids into cellulose-MOF composites. Expanding research in this direction could open the way towards novel cellulose-based materials with enhanced properties for various water treatment applications.

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