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**Review Paper**

# Biopolymer-based hydrogels for the remediation of heavy metals and dyes from wastewater

**Lina F.D. Al-Heety**[1*], **Omar M. Hasan**[2], **Khalil T. Hassan**[3,4]

[1]Department of Biotechnology, College of Science, University of Anbar, Anbar, Iraq
[2]Department of Biology, College of Science, University of Anbar, Ramadi, Iraq
[3]Department of Physics, College of Science, University of Anbar, Ramadi, Iraq
[4]Nanomaterials Research Center, University of Anbar, Ramadi, Iraq

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**Corresponding author**
*Lina F.D. Al-Heety*
*lena.fahmi@uoanbar.edu.iq*

## ABSTRACT

The increasing accumulation of heavy metals in aquatic environments poses a persistent threat to both environmental sustainability and public health. Conventional water treatment technologies often suffer from drawbacks such as high operational cost, limited selectivity, and poor regeneration efficiency. In recent years, biopolymer-based hydrogels have gained attention as efficient, eco-friendly, and tunable adsorbents for the removal of heavy metal ions from aqueous systems, owing to their high-water retention capacity, surface functional versatility, biodegradability, and environmental compatibility. Despite significant research efforts, current findings remain scattered, and the lack of a unified framework for evaluating hydrogel performance presents a challenge for practical implementation. This review provides a comprehensive and critical synthesis of recent progress in the design, functionalization, and application of natural biopolymers, particularly cellulose, starch, and chitosan, in hydrogel fabrication for water purification. Special emphasis is placed on chemical modification strategies, including carboxymethylation, graft copolymerization, and sulfonation, that introduce or enhance reactive functional groups responsible for metal ion binding. The mechanisms underlying metal ion removal, such as ion exchange, electrostatic interaction, coordination, and chelation, are discussed in detail. The role of crosslinking methods in controlling hydrogel structure, porosity, stability, and recyclability is also addressed. In addition, the integration of nanomaterials and biochar into hydrogel matrices for performance enhancement is highlighted. Finally, the review outlines key challenges and future directions in translating these materials from laboratory-scale studies to practical, scalable, and sustainable water treatment solutions.

**Keywords:** *Biopolymer hydrogels, dyes, Heavy metals, Polysaccharides, Water treatment*

## 1 INTRODUCTION

Water is the most abundant natural resource on Earth. However, only 3% of current reserves are freshwater; even less is available for daily use [1, 2]. Industries such as mining, tanneries, and metal plating significantly contribute to the pollution of the environment with harmful heavy metals [3]. Furthermore, natural resources and diverse human activities, sometimes known as anthropogenic activities, are the main sources of heavy metal releases into ecosystems. Heavy metals can naturally be released by volcanic eruptions, soil degradation (such as surface erosion), and crumbling rocks [4, 5].

Heavy metals can enter the human body through the skin, inhalation, or ingestion. Since the body lacks the ability to metabolize these substances, they accumulate in soft tissues, leading to considerable health hazards [6–8]. For example, regular ingestion of arsenic from drinking water can lead to human malignancies of the kidney, bladder, skin, and lungs [9]. Jomova et al. [10] stated that there is no denying the detrimental impact of several heavy metals on human health; their extended half-life in the environment is attributed to their resistance to natural breakdown. They can transform into insoluble or distinct compounds, harming ecosystems and human health. This phenomenon is explained in Figure 1, which shows types of heavy metal poisoning and their potential hazards to human health. One of the main challenges in today's society is the complete removal of organic and inorganic colors and heavy metal ions from wastewater, purifying it without harming the environment [11].

Several recently published articles have shown that many developed countries are currently evaluating treatment methods and techniques. Those include membrane filtration, microbial bioremediation, advanced oxidation processes (AOPs), carbon nanotechnology, electrochemical methods, chemical precipitation, solvent evaporation, ion exchange, photocatalysis, and biosorption for the removal of heavy metals from water [12, 13]. Adsorption remains the most straightforward and cost-effective method for removing pollutants from wastewater, and even naturally occurring sorbent materials have attracted significant interest [14].

Recently, researchers have shifted their primary focus to the design and development of novel polymeric adsorption matrices that could provide an affordable and effective treatment technology. From this perspective, hydrogels (HGs) are emerging as a more effective adsorbent for the treatment of various aqueous contaminants. They are insoluble, hydrophilic, cross-linked polymers that possess a high swelling capacity, enabling them to absorb a significant amount of water into their three-dimensional reticulated networks. A growing body of research has been published on HG engineering and its application to wastewater treatment with magnetic particles, nanoparticles, and other chemical catalysts [15, 16].

Hydrogel (HG) is a three-dimensional polymer network composed of hydrophilic functional groups such as –$NH_2$, -COOH, and others, which confer a high water retention capability and allow for the adsorption of various metal ions and dyes from wastewater [17]. In recent developments, hydrogels have also served as precursors for the synthesis of aerogels, which are produced via drying techniques (e.g., supercritical drying) that preserve their porous structure. Aerogels exhibit distinct physical properties, including extremely low densities (0.0001 to 0.2 g/$cm^3$), high specific surface areas (>200 $m^2$/g), and porosity exceeding 90% with dominant mesopores (2-50 nm) [18]. These structural features render aerogels attractive for environmental remediation applications, complementing the conventional use of hydrogels.

Hydrogels have attracted considerable attention because of their exceptional strength and diverse composite methodologies. It is essential to note that more sites participate in the adsorption process due to the abundant active functional groups on the gel polymer framework and its three-dimensional network structure. Due to these advantages, gels are commonly used as adsorbents in environmental protection. The presence of many functional groups (including -OH, -COOH, -$NH_2$, and -$SO_3H$) in hydrogels facilitates the adsorption, ion exchange, and chelation of heavy metal ions, which are chiefly responsible for their elimination. The benefits of fast adsorption, substantial adsorption capacity, and recyclability are evidenced by the adsorption of heavy metal ions on hydrogels, which have recently acquired prominence [19, 20].

Although recent literature explores a wide array of biopolymers, such as chitosan, alginate, and pectin, for wastewater treatment applications, this review specifically emphasizes cellulose and starch. This focus is justified by their abundance, biodegradability, renewability, and chemical versatility, which collectively make them ideal candidates for the development of sustainable hydrogel-based sorbents.

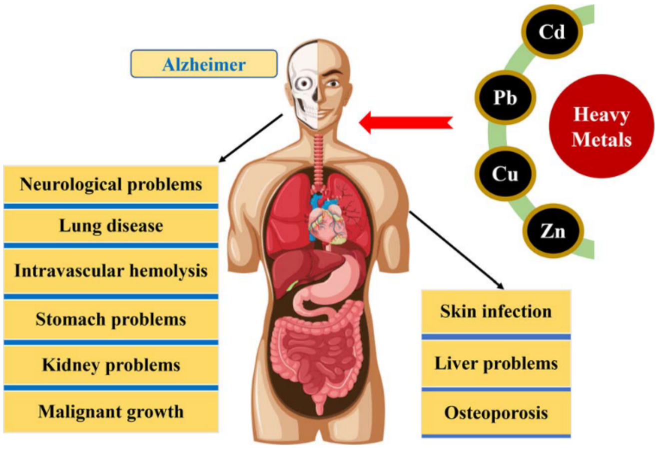

**Fig. 1** The effect of heavy metals on the health and safety of the human body [21]

## 2 HYDROGELS

Aerogels are generally defined as low-density, highly porous materials, primarily mesoporous, that are derived from sol-gel processes followed by drying techniques that preserve the internal network structure. These materials exhibit exceptional properties, including high surface area, open porosity, and ultralight weight, making them attractive for a range of high-value applications. The concept of aerogels dates back to 1931, when Samuel Kistler pioneered their fabrication using biopolymers. He demonstrated that natural polysaccharides such as gelatin, agar, nitrocellulose, and cellulose could serve as gel precursors, laying the foundation for the development of bio-based aerogels [22].

Additionally, hydrogels can also be classified based on the source [23]. They are classified as either interpenetrating, multipolymer, or homopolymer polymers based on the polymer components they contain. While co-polymers are composed of two or more different types of monomers with a single hydrophilic moiety, homopolymers are formed from a single type of monomer. A notable class of hydrogels known as interpenetrating polymers is composed of two self-reliant, cross-linked synthetic or natural polymer units [24].

The water-absorption capacity of a hydrogel is determined by covalent bonds, influenced by the polymer's hydrophilic nature and the degree of reticulation in the matrix. Physical hydrogels form through molecular entanglements, physical contact, ionic bonding, or hydrogen bonding, and they can be reversibly switched. Consequently, these materials are also known as temporal hydrogels. Hydrogels are classified into three categories based on charge: cationic, anionic, and neutral [25].

Hydrogels possess a three-dimensional polymeric network that can entrap heavy metal ions within their porous structure, as shown in Figure 2. This architecture enables strong physical and chemical interactions with pollutants via mechanisms such as electrostatic attraction and chelation. The bound water within the hydrogel provides additional active sites, facilitates hydrogen bonding, and enables the diffusion of tiny hydrophilic molecules. Many polar functional groups, such as hydroxyl (-OH), amino (-$NH_2$), and sulfonyl (-$SO_3H$), are integrated into the polymer chains, further enhancing the adsorption potential. These chains interact via van der Waals forces, hydrogen bonds, and covalent linkages, resulting in high water retention and structural stability. This unique combination of features enables hydrogels to efficiently capture and immobilize metal ions in aqueous environments [26].

Swelling behavior is a critical functional property of hydrogels. The swelling and deswelling behavior of hydrogels plays a critical role in determining their performance in real-world applications. In drug delivery systems, for instance, swelling governs the rate and timing of drug release, which can be modulated by environmental triggers such as pH or temperature. Similarly, in the context of pollutant uptake, the swelling capacity affects the diffusion of contaminants into the polymer matrix and their subsequent adsorption onto functional groups. Therefore, swelling/deswelling tests are essential for evaluating the hydrogel's responsiveness, stability, and functional efficiency under varying environmental conditions [27].

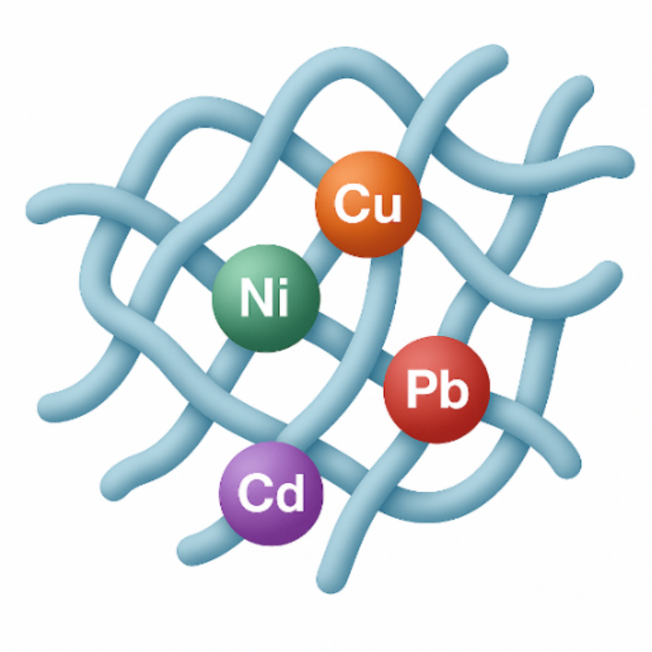

**Fig. 2** Schematic diagram showing the retention of heavy metal ions within a polymeric hydrogel network

The limited solubility of native biopolymers, such as cellulose and starch, in conventional solvents poses a significant challenge for the synthesis of hydrogel systems. This is primarily due to their high crystallinity and extensive intermolecular hydrogen bonding, which restrict their dissolution in water and most organic or inorganic solvents [28].

While native cellulose is indeed insoluble in most conventional solvents due to its high crystallinity and extensive hydrogen bonding, recent advances have introduced more effective strategies for its dissolution and for the formulation of hydrogels. Among these, deep eutectic solvents (DESs), which are green, biodegradable alternatives, have attracted attention for their ability to disrupt cellulose's hydrogen-bonding network and enable homogeneous processing.

Furthermore, nanoscale crosslinking techniques, such as UV-induced or chemical crosslinking using multifunctional agents, have been employed to enhance the structural integrity and performance of cellulose-based hydrogels (CBHs). These modern approaches significantly expand the toolbox for cellulose functionalization and hydrogel fabrication beyond traditional solvent systems like LiCl/DMAc or ionic liquids [29]. The practical efficacy of hydrogel-based systems in removing heavy metals and organic pollutants from wastewater is supported by a wide range of studies, as summarized in Table 1.

**Table 1** Bio-based composites for the effective removal of pollutants in wastewaters

| **Hydrogel** | **Pollutant** | **Removal time (min)** | **pH** | **Adsorption capacity** | **References** |
|---|---|---|---|---|---|
| Starch/activated carbon composite | Methylene blue | 90 | 10.5 | 90% | [30] |
| Carboxymethyl cellulose | $Cd^{2+}$, $Pb^{2+}$ | – | 4.0 | 540, 810 mg $g^{-1}$ | [31] |
| Starch/Fe-Mn binary oxide | $As^{5+}$ and $As^{3+}$ | 120 | 7 | 160.63 and 284.64 mg $g^{-1}$ | [32] |
| Chitosan | $Zn^{2+}$, $Cu^{2+}$ | – | – | 50%, 80% | [33] |
| Polyethyleneimine functionalized chitosan-Lignin | $Hg^{2+}$ | 120 | 5 | 663.5 mg $g^{-1}$ | [34] |
| Carboxymethyl cellulose-graft-poly (acrylic acid) hydrogel | $Ni^{2+}$ | – | – | 366.11 mg $g^{-1}$ | [35] |
| CMC-HEC hydrogel | Cd, Methylene blue | 60<br>60 | 7<br>9 | 126.58 mg $g^{-1}$<br>69.23 mg $g^{-1}$ | [36, 37] |
| Cellulose/ chitosan | $Cu^{2+}$ | – | 5 | – | [38] |
| Cellulose | dyes | – | – | 84% | [39] |
| CS/CMC-PEG hydrogel | Methylene blue | 30 | 11 | 331.72 mg $g^{-1}$ | [40] |

## 2.1 Cellulose

Cellulose is composed of $\beta\,(1 \rightarrow 4)$ glycoside linkages, Figure 3, which join the sole D-glucose units (300-3000) to one another. For example, plants (natural polymers) require cellulose to provide them with stiffness and strength. Wood has roughly 50% cellulose and cotton contains 95% pure cellulose. In contrast to other naturally occurring polysaccharides, cellulose is the most widely distributed kind on Earth. Because hydroxyl groups and oxygen atoms on adjacent chains form intermolecular hydrogen bonds, cellulose is one of the most robust and long-lasting natural materials. Cellulose has more crystalline lattices and is less soluble in water than chitosan. Nevertheless, unaltered cellulose lacks certain qualities that make it useful since it lacks strong metal ion removal ability and varying physical stabilities serving as a powerful adsorbent [41].

Heavy metal ions in water can be effectively adsorbed by the surface functional groups of cellulose-based hydrogels. While some CBHs exhibit large surface areas and high porosity. These properties are highly dependent on the preparation method and crosslinking strategy employed, which influence the internal network structure and accessibility of active sites. Cellulose-based hydrogels can be functionally enhanced to selectively adsorb heavy metal ions by grafting specific chelating groups onto their polymer backbone. For example, thiol (-SH) groups exhibit a strong affinity toward soft heavy metals such as mercury ($Hg^{2+}$) and lead ($Pb^{2+}$), whereas amine ($-NH_2$) and carboxyl (-COOH) groups are more effective in binding with harder metal ions like copper ($Cu^{2+}$), cadmium ($Cd^{2+}$), and zinc ($Zn^{2+}$). These chemical modifications improve the sorption capacity, selectivity, and reusability of the hydrogel under various environmental conditions [25]. The three-dimensional materials with pore structures that facilitate super water absorption, water retention, and specialized swelling capabilities are hydrogels manufactured of nanocellulose through physical and chemical crosslinking. Low-cost, non-toxic materials, such as modified lignocellulosic materials derived from renewable biological resources, have been developed and are being used for a variety of purposes because they are biodegradable and sustainable. Carboxymethyl cellulose (CMC) is a cellulose derivative in which carboxymethyl groups ($-CH_2-COOH$) are introduced into the polymer backbone. The presence of these carboxyl functional groups imparts high hydrophilicity and water solubility to CMC, enabling the formation of hydrogels with excellent swelling capacity. Moreover, the ionizable nature of the carboxyl groups makes CMC-based hydrogels sensitive to pH and ionic strength, which is advantageous for applications such as controlled drug release and pollutant adsorption [42].

Godiya et al. [43] developed a composite hydrogel based on carboxymethyl cellulose and polyacrylamide for the removal of Cu, Cd, and Pb ions from aqueous solutions. The highest adsorption efficiencies were 227.2 mg g-1 for Cu, 256.4 mg g-1 for Cd, and 312.5 mg g-1 for Pb. Adsorption experimental data followed the pseudo-second-order kinetics and the Langmuir isotherm model. Furthermore, cellulose has a large number of hydroxyl groups, making it a useful material for the production of adsorbents that remove heavy metal ions from aqueous media. Wang et al, (2017) created a bio-adsorbent that was supported by sugarcane cellulose and used it to remove heavy metals. A cellulose-supported bio-adsorbent was used to remove Cu(II), Zn(II), and Pb(II) metal ions; the maximum adsorption capacities were reported to be 446.2, 363.3, and 558.9 mg $g^{-1}$, respectively. The produced bio-adsorbent was incredibly effective and environmentally sustainable [44].

Wu et al. [45] prepared A cellulose-based hyper branched adsorbent (MCC/HBPA-0.88) via cross-linking with microcrystalline cellulose (MCC) and amino-terminated hyper branched polymers (HBP-NH2). Hyperbranched polymers are highly branched, three-dimensional macromolecules with a large number of terminal functional groups, such as amines ($-NH_2$). Their structure allows for a high density of reactive sites, which enhances the interaction capacity with metal ions or dyes, making them particularly effective for adsorption applications. This strategy for liquid-solid phase one-step rapid conversion could effectively increase the conversion rate of the reactants (above 99%). [46] reported that a cellulose-supported bioadsorbent derived from hemp fiber, rich in biomacromolecules such as cellulose and hemicellulose, exhibited enhanced adsorption capacity for anionic dyes at elevated temperatures and under acidic pH conditions. More recently, a sulfated cellulose–magnetic biocomposite (MSHB) synthesized from hemp biomass demonstrated high removal efficiency for cationic dyes across a wide pH range, highlighting the versatility of hemp-derived materials in wastewater treatment applications [47].

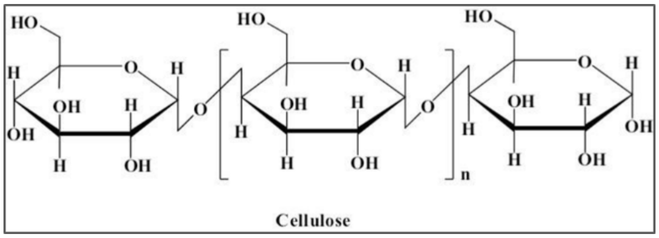

**Fig. 3** Structure of Cellulose

## 2.2 Modification of cellulose

Modified cellulose, such as that produced through chemical crosslinking or carboxymethylation, is widely used as a raw material for the synthesis of cellulose-based hydrogels [48]. Three alcoholic hydroxyl groups are present in each hydroxyl-glucopyranose unit of cellulose molecules, allowing possible chemical modification of these hydroxyl groups. There are 30 to 36 cellulose chains in crystalline areas that are connected by hydrogen bonds (MC). These chains often cross both crystalline and amorphous areas. Methylcellulose [49]. Methylcellulose is classified as a thermoresponsive polymer with a lower critical solution temperature (LCST), meaning it undergoes a sol–gel transition upon heating. This reversible thermal behavior enables the hydrogel to form a gel at elevated temperatures and return to a sol upon cooling, which is particularly useful for controlled drug delivery, temperature-triggered pollutant release or capture, and recyclable adsorption systems. The LCST property allows the hydrogel's swelling and binding characteristics to be modulated by temperature, offering tunable performance in dynamic environmental conditions [50]. In addition, chemical cross-linking is required to increase the adsorbent's active sites and wet mechanical characteristics to enhance its adsorption performance. Accordingly, a variety of modified cellulose adsorbents have been recently used to remove heavy metal ions from wastewater [20,51,52].

The modification process of carboxymethylating nanocellulose entails adding carboxymethyl groups to the surface of the fibers. This modification modifies the chemical and physical characteristics of the material, increasing its water solubility and improving its surface charge. Furthermore, the application field is further expanded by physical or chemical cross-linking techniques, blending preparation, polyelectrolyte complex synthesis, and interpenetrating polymer network technology [53]. Kundu et al [54] fabricated a new composite hydrogel, made of carboxymethyl cellulose (CMC), microcrystalline cellulose, and xylan, in an alkaline medium. They used diglycidyl glycol ether as a crosslinking agent, which enabled it to adsorb Cd2+ and divalent nickel ions Ni2+ from water. The hydrogel could absorb a maximum of 55.85 mg/g and 61.44 mg/g of the adsorbent.

## 2.3 Starch

Starch, a low-cost polysaccharide having a molecular formula of $(C_6H_{10}O_5)$ n. As seen in Figure 4, the two distinct polymer chains that make up starch are amylose and amylopectin [55]. Amylose is a mostly linear polymer composed of $\alpha(1 \rightarrow 4)$-glycosidic linkages, while amylopectin is a highly branched polysaccharide that contains both $\alpha(1 \rightarrow 4)$-glycosidic bonds along its linear chains and $\alpha(1 \rightarrow 6)$-glycosidic linkages at the branching points. The structural differences between amylose and amylopectin, primarily in their glycosidic linkages and degree of branching, result in distinct physicochemical properties, including differences in solubility, gelatinization behavior, and interactions with other molecules. For example, the short branching of amylopectin at the 1, 6-glycosidic connections is responsible for the crystalline area of the granules [56, 57]. Turning starch into gelatin includes three main stages. Hydrophilic starch granules first expand after absorbing water. The granule structure breaks down when the starch dissolves in water and transforms into a gelatin in the subsequent stage. The final stage involves restructuring the polysaccharide structure and forming the starch hydrogel network through cooling and aging. The retrogradation phase is another name for this stage [58, 59].

Hydrogels based on starch can be prepared via various chemical methods, such as etherification and starch grafting. When starch molecules are etherified, other ether groups, such as carboxymethyl starch, replace the -OH groups on the starch molecule. On the other hand, the grafted starch method produces hydrogels by joining different vinyl monomers with starch [60, 61]. The potential application of this hydrogel in wastewater treatment is indicated by its capacity to absorb both cationic and anionic dyes. Catalytic starch-containing ammonium groups were created by combining starch with 3-chloro-2-hydroxypropyl trimethyl) ammonium chloride. These groups were then utilized to create starch-derived Nanocomposites for the adsorption of cationic dyes [62].

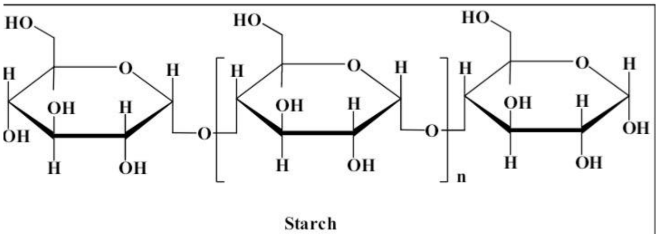

**Fig. 4** Structure of Starch

## 2.4 Modification of starch

The term "rich modification" describes initiatives to boost the synthesis of starch while simultaneously altering its composition, structure, and introducing necessary components to meet particular end objectives [63]. Starch is a natural hydrophilic polysaccharide consisting of linear (amylose) and branched (amylopectin) glucose polymers. It has outstanding properties, including non-toxicity, biodegradability, biocompatibility, and low cost, owing to its high surface hydroxyl groups. However, natural starch faces some major problems that make its direct use as an adsorbent for wastewater treatment difficult. These include its small surface area, poor solubility in water, low molecular weight, instability at high temperatures, and lack of reactive functional groups [64].

Guo et al. [65] investigated cross-linked cationic starch for removing the golden yellow color from aqueous solutions. The proposed cross-linked cationic starch was synthesized using corn starch as raw materials, 3-chloro-2-hydroxypropyl trimethylammonium chloride for cationic etherification, and epichlorohydrin as a cross-linked agent, resulting in a maximum adsorption capacity of approximately 208.77 mg/g at 308.15 K. High-pressure processing (HPP) is a non-thermal technique primarily applied in the food industry, where starch-containing materials are subjected to pressures ranging from 100 to 600 MPa at room temperature for short durations (2–30 minutes). While HPP can alter the gelatinization and retrogradation behavior of starch, its application in adsorbent or environmental treatment contexts remains limited and relatively unexplored compared to conventional chemical or enzymatic modification methods. This process inactivates germs and enzymes and modifies the structure of food constituents, such as starch [66]. Also, pulsed electric field, applying high-voltage electrical pulses (1–80 kV/cm) to starch pastes or suspensions for brief periods (micro to milliseconds), is known as PEF modification of starch. As a result of this process, the starch molecules undergo both chemical and physical modifications, breaking down into smaller starch granules and forming cross-links. Ultrasound processing (USP) is a non-thermal technique that uses high-frequency sound waves to induce cavitation, which can disrupt granular starch structures. This process increases surface area, introduces microporosity, and may lead to partial depolymerization or exposure of hydroxyl and carboxyl groups, thereby increasing the number of active adsorption sites. While USP has been widely used in food processing, its role in modifying starch for environmental remediation applications—such as the removal of dyes or heavy metals—is gaining attention due to its ability to tailor starch's structural and chemical features. Utilizing USP, starch modification is often implemented in liquid systems where starch suspensions experience brief cycles of compression and expansion, also known as the cavitation effect [67].

Investigated modifying starch using extrusion The researchers noticed a more profound reduced solubility and phase transition heat, greater resistance to the action of amylolytic enzymes, and physical and chemical color changes could all lead to changes in non-digestible carbohydrates. They presented their technique as a means of getting over starch resistance. In an experiment conducted by Cabrera-Ramirez and colleagues [68]. With the chemical modification, a greater affinity can be noted between the adsorbents and the surface of the adsorbates. Therefore, modified quinoa starch particles containing dodecanoyl chloride (MQS) were produced to adsorb cationic compounds, oils, and heavy metals such as cadmium, mercury, and lead. The resulting emulsion proved effective in removing oils (92%), reducing turbidity (98%), and reducing chemical oxygen demand (COD) (87%). Heavy metal removal was even more efficient, with an average removal rate of 85% for metals used in industrial wastewater [69]. In the one-pot production technique, carboxymethyl starch (CCMS) and methacrylic acid (MAA) can also be grafted and micro-cross-linked to make (CCMS-g-MAA). The CCMS-g-MAA was able to get rid of a lot of Pb (II) (57.13 mg/g at pH 4) and Zn (II) (51.41 mg/g at pH 5) when the sample dose was 0.68 g/L [70].

### 2.5 Chitosan

The Greek word "Chiton," meaning "covering" or "envelope," is the source of the English term "chitin." [71]. Chitosan, along with cellulose, is among the most widely studied natural biopolymers, particularly because of its nitrogen-containing structure derived from chitin. N-deacetylated chitin Figure 5, which can be found in crustacean shells and insect cuticles, is the basic component of chitosan. It is an example of a biodegradable, renewable glucose distinguished by its linear amino polysaccharide. Chitin deacetylase catalyzes the N-deacetylation of acetylated glucosamine units, facilitating the bioconversion of chitin into chitosan. To be classified as chitosan, the deacetylation degree must typically exceed 50%, which alters the polymer's solubility, charge density, and reactivity, making it suitable for applications such as adsorption, drug delivery, and biomedicine. Chitosan exhibits amphiphilic behavior, possessing both hydrophilic and hydrophobic characteristics. Its overall affinity to water depends on environmental factors such as pH and degree of deacetylation. In acidic media, chitosan becomes protonated and hydrophilic, allowing it to dissolve and interact with aqueous-phase species. However, under neutral or basic conditions, it may display hydrophobic behavior due to reduced solubility and stronger inter- and intramolecular interactions. This tunable duality enhances its sorption capacity and makes it suitable for a wide range of chemical and functional modifications. It has been extensively investigated for applications in agriculture and industry, including its use as a biocatalyst in wastewater treatment, drug or gene delivery, and cell/enzyme immobilization [72, 73].

This hydrogel has a porous structure, which means it has a greater specific surface area and more binding sites. It can also adsorb metal ions such as $Pb^{2+}$, $Cu^{2+}$, and $Cd^{2+}$, demonstrating its ability to adsorb heavy metal ions. When making chitosan-based hydrogels, the interactions between chitosan molecules, whether they are between molecules or inside molecules, must be strong enough to create physical binding sites. In an alkaline environment, hydrogen bonding between chitosan molecular chains is enhanced, leading to microcrystalline structures that act as physical binding sites. The substantial salting effect of highly soluble halide salts such as calcium chloride, potassium chloride, and magnesium chloride in the chitosan solution strengthens the hydrophobic interactions between chitosan molecules. The physical binding points result from molecular chain aggregation, leading to physical entanglement. N-glucosamine forms ligand interactions with polyvalent anions, creating an ionic crosslinked network in which ionic ligand bonds serve as the physical binding sites. Alternatively, polyvalent anionic salts, such as sulfate and citrate, may be included in the chitosan solution to dehydrate the chitosan molecules. The amino and hydroxyl groups of chitosan facilitate both chemical and physical crosslinking, providing numerous benefits including cost-effectiveness, renewable resource accessibility, biocompatibility, biodegradability, and multifunctionality [74].

A composite that included hydroxyapatite and chitosan, among other ingredients, shows that at room temperature it can extract 50.39% and 74.77% of $Zn^{2+}$ and $Cu^{2+}$ ions, respectively, from an aqueous solution. Recent years have seen a significant increase in interest in chitosan as a biopolymer hydrogel raw material due to its widespread availability, excellent biocompatibility, strong adsorption capacity, biodegradability, and non-toxic breakdown products. Chitosan-based hydrogels and their derivatives are essential in the removal of heavy metal ions because they possess a wide variety of active amino and hydroxyl groups that can dissociate cations, and form chelates with metallic ions. To adsorb Co ions from solutions, Zhuang and colleagues synthesized chitosan-g-maleic acid using gamma radiation; the chitosan adsorption limit was 2.78 mg/g [75].

Tang et al. [76] used a physical crosslinking technique to create a new chitosan-based double network hydrogel. This hydrogel has the highest removal capacity of 176.50 mg/g demonstrating its superior impact on the adsorption of heavy metal ions including $Pb^{2+}$, $Cu^{2+}$, and $Cd^{2+}$. An improvement in the simultaneous adsorption capacity of chitosan for metal ions can be achieved by grafting with mono carbamoylcarboxylic acids using a coagulation-flocculation process. The grafting of chitosan with carbamoylcarboxylic acids changes its physicochemical properties as solubility and porosity, limits of potential and isoelectric point [77] . In the same way, carboxymethylation enhances the active sites on the chitosan surface, acidic media stability, and enhanced multi-heavy metal adsorption-desorption and subsequent regeneration potential. The excellent functionality of the carboxymethyl-chitosan resin enhances the simultaneous removal of metal ions from real-world effluent systems [78].

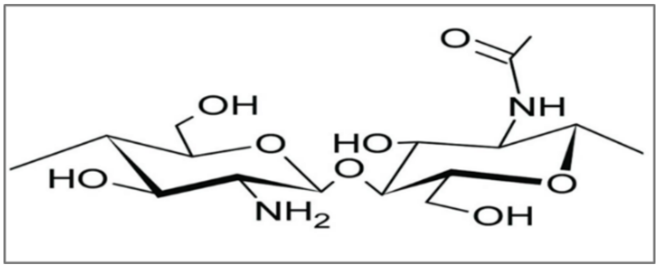

**Fig. 5** Structure of Chitosan

## 3 MECHANISM OF HEAVY METAL IONS REMOVAL BY HGS

Numerous industries, including the extraction of minerals, water treatment, metal molding, metal coating, batteries, nuclear industry, leather tanning, electroplating, fuel combustion, landfills, electronics, metal finishing, wood processing, enameling, and nuclear power generation release heavy metals into water sources [79–82]. Heavy metals are considered one of the toxic pollutants in industrial wastewater and are of great concern due to their negative impact on human health and ecosystems [51,83]. The adsorption approach employing hydrogels (HGs), as illustrated in Figure 6, is considered among the most effective strategies for the removal of various heavy metals (HMs) from contaminated water, owing to their low environmental footprint, and high removal performance. Especially in low-concentration systems, due to the sorbents' reusability and low cost.

To further improve and adjust HG properties, a thorough understanding of the adsorption process and the removal mechanisms of various pollutants on individual HGs is crucial. The characterization of the sorption process of metal ions is a complex phenomenon that requires a thorough understanding of process parameters. Interactions such as ion exchange, complexation, electrostatic interaction, hydrogen bonding, hydrophobic interaction, acid-base interactions, coordination/chelation, and so on occur depending on the various functional groups of HGs, pollutant chemistry, and experimental conditions (pH, salt concentrations, temperature, ligands, ionic strength, and contact time) [84].

Hydrogels possess a porous three-dimensional (3D) polymeric network that enables efficient absorption and retention of water and dissolved substances, including heavy metal ions [85, 86]. Their high porosity and swelling capacity allow pollutants to diffuse into the internal structure, where they interact with chemically reactive functional groups [87,88]. These include hydroxyl, carboxyl, amino, and other polar groups that facilitate binding through electrostatic attraction, coordination, and chelation [89,90].

Due to these structural and chemical characteristics, hydrogels are gaining increasing attention for their effectiveness in removing heavy metals from wastewater, particularly in systems that require low-cost, efficient, and reusable sorbents [91]. Hydrogels are classified as hydrophilic gels, and their ability to respond to environmental stimuli such as pH and ionic strength further enhances their versatility in water treatment applications [92].

The sorption process typically involves a liquid phase (containing dissolved heavy metal ions) and a solid hydrogel phase (the sorbent), in which metal ions interact with functional groups on the hydrogel matrix via a combination of physical and chemical mechanisms [93]. It is a complex process involving chemisorption, complexation, adsorption on surfaces and in pores, ion exchange, chelation, adsorption by physical forces, and entrapment in inter- and intrafibrillar capillaries and spaces of the structural polysaccharides network as a result of the concentration gradient and diffusion through the cell wall and membrane is responsible for the sorbent's high affinity for metal ions [94].

There are a variety of functional groups on sorbent surfaces that can bind heavy metal ions. These include oxygen-containing groups (e.g., hydroxyl -OH and carboxyl -COOH), acetamide, carbonyl, phenolic, amido, amino, sulphydryl, structural polysaccharides, and esters. A negatively charged sorbent surface is necessary for the adsorption of positively charged metal ions from model solutions in water-based systems. There is a connection between these groups and metal complexation. Certain biosorbents are specific for particular types of metals based on their chemical makeup, while others are non-selective and bind to a wide variety of heavy metals without any particular priority [95].

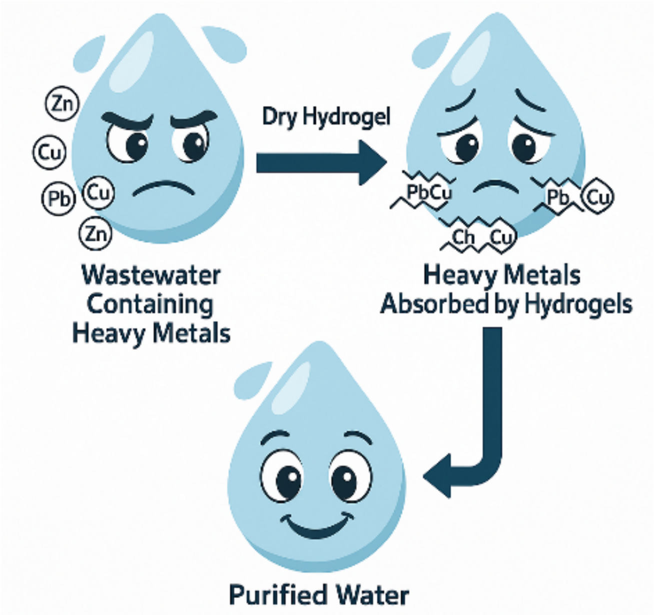

**Fig. 6** Schematic illustration for the removal of heavy metals from wastewater by using a hydrogel

### 3.1 Ion exchange approach

Ion exchange is the process of moving ions from an insoluble material to a liquid (water). Negatively or positively charged ions that are undesirably soluble are removed from an aqueous solution. The total number of charged particles released from the outermost layer of the adsorbent and the number of ions absorbed by the adsorbent particles are equal in a perfect ion exchange process [96]. Ion exchange is a highly practical and effective method, particularly for removing hazardous chemicals from wastewater, such as heavy metals and dyes. This procedure reduces the level of hazardous load by changing the form of pollutants so they can be recycled, replacing harmful elements with less hazardous ones, or by facilitating eventual disposal by reducing the hydraulic flow of the toxic element-carrying stream [97].

pH is very important in the adsorption process because it affects the solubility of metal ions in water and the ionization state of the amino groups in the bioadsorbent, which are what hold heavy metals together. As you can see, the amount of metal removed increased significantly at higher pH levels. This could be because there aren't enough protons, which could be competing with ionic metals for the chelation with amino and carbonyl groups. As the pH rises, more amino groups will become less ionized and more likely to bind with positively charged metal ions. However, at higher pH values, the lower binding capability was because the metallic ions were less soluble, [98].

### 3.2 Electrostatic interaction

The association of charged units is known as electrostatic interaction, and it can happen when molecules are oppositely charged (as in action-anion contacts) or identically charged (as in cation-cation or anion-anion interactions). For a cellulose-based hydrogel to eliminate ionic pollutants by electrostatic attraction, each ion that needs to be adsorbed must have a charge opposite to the adsorbent's surface. To enable the cellulose-based hydrogel to deliver an opposing charge to the required ions, certain functional groups are added, depending on the kind and chemical composition of the pollutants [99].

Several investigations have proposed that electrostatic forces induce the adsorption of ionic dyes and heavy metals in different HGs [43,97]. Favorable electrostatic interactions were used to crosslink HGs based on nanomaterial hybridization. These HGs show remarkable capacities for self-revival on their own, bestowing reusable traits [100]. Based on this, in order to remove organic dyes such as methylene blue (MB) and rhodamine B (RhB), Guo et al. [101] prepared GO/polyethylene mine (PEI) HGs with excellent dye adsorption capacity. The dye adsorption capacity of the hydrogel is mainly attributed to the GO sheets, whereas the PEI was incorporated to facilitate the gelation process of GO sheets. In a related study, TiO2 NPs and AA-grafted alginate were combined to create HG nanocomposites intended to remove the cationic dye methylene blue (MB). With increasing acrylamide (AA) dosage, the gel material's maximum sorption capacity increased to 99.4%. It was suggested that the electrostatic interactions between e-COOH moieties on HGs and MB (cationic ions) were the cause of the association complex. There have also been reports of secondary interactions via H-bonding between the e-OH of HGs and the imine groups e-RCH-NR of dye [102].

### 3.3 Adsorption approach

Adsorption is the process by which molecules of a substance accumulate at a surface, resulting in a higher concentration compared to the bulk phase. The phenomenon relies on the existence of an adsorbent layer capable of accommodating a micro-substance on its surface, characterized by a specific surface structure [103, 104]. Adsorption-based techniques are generally considered operationally simple, cost-effective, and efficient for removing heavy metals from wastewater. However, different types of adsorbents require distinct reactivation or regeneration methods, which can significantly influence the overall operational cost and sustainability of the treatment process [105]. Adsorbents do not produce any additional harmful compounds after exposure to toxic pollution. Adsorption-based procedures are widely employed in the water treatment industry and are thought to be among the most economical ways to eliminate heavy metal ions [45].

The ability of adsorption to eliminate heavy metals at lower concentrations, together with its low energy consumption and availability of raw materials, renders it an effective method. Physisorption and chemisorption are the two primary types of adsorptions. Van der Waals forces bind the absorbent and absorbate in physisorption, while chemical bonds are established between them in chemisorption as shown in Figure 7 [106]. To further improve Cellulose-Based Hydrogels (CBHs) to boost their adsorption efficiency, a full understanding of the adsorption process and the removal mechanisms of diverse pollutants on individual CBHs is extremely required. CBH adsorption usually occurs through a variety of interactions. Their nature depends greatly on the functional groups found in the HG, the characteristics of the adsorbent, the chemical makeup of the pollutants, and the parameters of the experiment (e.g., concentration of the pollutant at the beginning, pH of the solution temperature, coexistence of metal ions, etc.) [99].

Hydrogels' molecular structure includes functional groups like amino, carboxyl, and hydroxyl, which increases their adsorption ability and makes them an excellent adsorbent for pollutant removal [107]. Numerous hydrophilic groups found in the hydrogel skeleton can bind with and immobilize metal ions. Additionally, the hydrogel's three-dimensional network structure can guarantee ensure the metal ions are evenly distributed in order to stop them from oxidizing and aggregating [108].

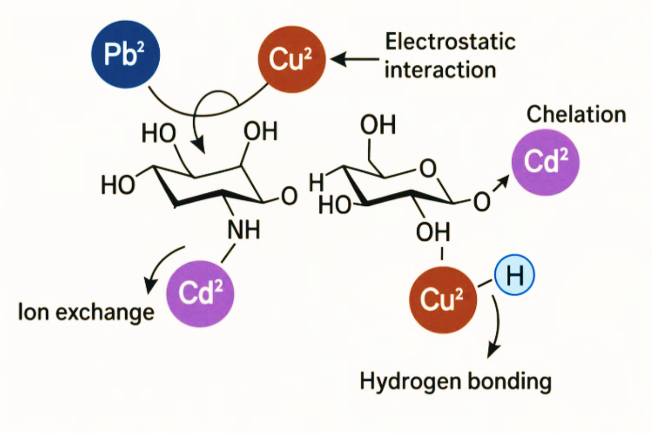

**Fig. 7** Representative mechanisms of heavy metal ion interactions with hydrogel functional groups

## 4 SYNTHESIS OF HYDROGELS BASED ON CROSSLINKING METHODS

A successful adsorption process begins with the production of an adequate super adsorbent, sometimes referred to as a hydrogel-derived adsorbent, it has a commendable ability to adsorb heavy metals. Three necessary molecules, a monomer, an initiator, and a cross-linker, are used to create the hydrogel. The following lists a few hydrogels, along with the initiators, crosslinkers, and monomers reported in various articles. The monomers of poly (2-acrylamido-2-methyl-1-propansulfonic acid-co-vinyl imidazole) hydrogel are N-vinyl imidazole and 2-acrylamido-2-methyl-1-propansulfonic acid (AMPS), which are crosslinked using N,N'-Methylenebisacrylamide (MBA) crosslinker. Initiating this reaction is 2,20-azobis (2-methyl propionamide, MPA) dihydrochloride [109].

In hydrogel–biochar composites, biochar primarily functions as an adsorbent enhancer and structural modifier, providing additional surface area, porosity, and functional groups that enhance overall adsorption performance. The hydrogel matrix is typically synthesized by polymerizing acrylamide (AAm) as the primary monomer and crosslinking it with N,N'-methylenebisacrylamide (MBA), which stabilizes the three-dimensional polymer network and enables uniform biochar integration. This process yields composites with enhanced mechanical strength, stability, and adsorption capacity [110].

As illustrated in Figure 8, hydrogel synthesis involves two fundamental elements: polymerization initiation and network crosslinking. Initiation can be achieved through various methods such as thermal initiation (e.g., using ammonium persulfate, APS), photochemical initiation (e.g., UV light with a photoinitiator), or redox initiation, each influencing the polymerization rate and final hydrogel structure. Crosslinking, in turn, occurs via physical interactions, such as hydrogen bonding, ionic interactions, and crystallization, or through chemical bonding with covalent crosslinkers, such as N,N'-methylenebisacrylamide (MBA). The choice between physical and chemical crosslinking determines key hydrogel properties, including mechanical strength, stability, porosity, and responsiveness to environmental stimuli [18, 29].

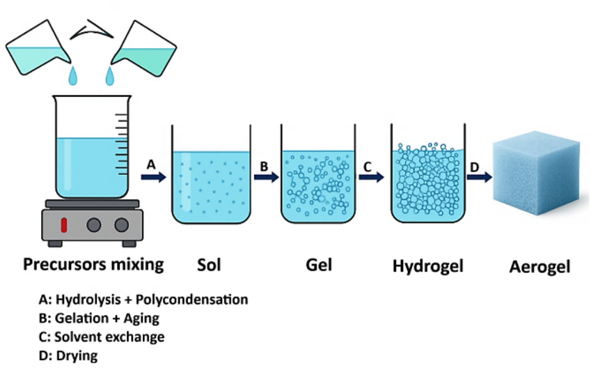

**Fig. 8** The schematic shows the synthesis stages of the hydrogel

### 4.1 Physical crosslinking

Non-covalent bonds exist between molecules and polymeric chains in physically cross-linked gels [111]. Physical hydrogels form through reversible, non-covalent interactions such as hydrogen bonding, ionic interactions, or hydrophobic interactions. While this allows for responsiveness to environmental stimuli, it also results in limited mechanical stability under stress or fluctuating conditions. As a result, physical hydrogels are generally less suitable for applications requiring long-term structural integrity, compared to chemically crosslinked hydrogels, which form stable covalent bonds and exhibit greater durability [112]. The hydrogels made with this method are also known as reversible hydrogels because they dissolve or melt at high temperatures and the resulting gel state vanishes when the conditions change. Because crosslinking agents can be avoided and physical crosslinks are easy to manufacture, structured hydrogels offer increased biological compatibility, minimal or absent toxicity, and ease of degradation, making them eco-friendly and sustainable [83]. Alginate hydrogels, for instance, are formed through ionic crosslinking with multivalent cations such as $Ca^{2+}$, $Ba^{2+}$, or $Al^{3+}$, which interact with the carboxylate groups on the alginate chains. The efficiency and mechanical strength of these hydrogels depend heavily on the valency and concentration of the crosslinking ions, higher-valency ions generally produce more stable and tightly crosslinked networks. In contrast, agarose gels are structured through physical crosslinking via hydrogen bonding between polymer chain segments. Additionally, nanocellulose-based hydrogels prepared by physical methods exhibit high porosity and water absorption, primarily due to the abundance of functional groups such as amino ($-NH_2$) and carboxyl ($-COOH$) on their surfaces [76].

Maiti et al. [113] found that the carbonyl group (C=O) of the incorporated guar oxide gum participated in the physical crosslinking process of a multicomponent chitosan-based hydrogel. The interaction was primarily attributed to hydrogen bonding between the carbonyl groups and protonated amino groups ($-NH_3^+$) on the chitosan backbone, which contributed to the formation and stabilization of the hydrogel network. The simplest and most straightforward method for creating hydrogels from natural materials is the freeze-thaw method. The process of thawing is precisely the opposite of freezing and is done at room temperature without the need for organic solvents. To prepare physical hydrogels, the freeze-thaw procedure is repeated 2 or more times. Materials primarily generated from cellulose are employed in this technique. Additionally, these hydrogels exhibit distinct swelling behavior in both soil and water [114].

### 4.2 Chemical crosslinking

Chemical crosslinking, which includes radiation polymerization, enzymatic polymerization, copolymerization, graft polymerization, and interpenetrating network crosslinking, is the creation of covalent chemical bonds in stable three-dimensional hydrogels [115]. By blending and crosslinking cellulose hydrogel with acrylamide and acrylic acid, Zhao et al. created a modified biosorbent. The modified hydrogel demonstrated maximum adsorption capacities of 157.51, 393.28, and 289.97 mg/g for divalent copper (Cu), lead (Pb), and cadmium (Cd) ions, respectively [116].

The physical crosslinking method often results in hydrogels with low stability and high dissolution rates; in contrast, the chemical crosslinking method increases hydrogel stability However, the use of chemical crosslinkers may raise concerns regarding environmental toxicity or biocompatibility, which must be considered in practical applications. Hu et al. [117] used sodium alginate (SA) and carboxymethyl cellulose (CMC) as core materials. The researchers used chemical cross-linking to create a more stable synthetic polymer outer layer, while the inner polysaccharide core was formed through physical cross-linking. This resulted in a bilayer hydrogel with distinct inner and outer compositions, enhancing the hydrogel's strength and stability while reducing swelling of the inner core and dispersion of its contents. Covalent bonds can be formed between polymeric chains via chemical cross-linking, which is produced by methods like Michael addition or free radical polymerization. In addition to providing stability, these cross-linking processes enable the hydrogel network to react to stimuli. For example, thermoresponsive hydrogels can be produced by adding temperature-sensitive cross-linkers, like N,N'-methylenebis(acrylamide) (MBAAm), to the polymerization mixture [118].

## 5 FUNCTIONALIZATION OF BIO-BASED MATERIALS

Biomaterials are a renewable, eco-friendly alternative, especially when they come from lignocellulose biomass. When paired with other activities, these biomaterials could be used for effective absorption and cleanup [119]. Functional biomaterials offer many benefits, including biodegradability, biocompatibility, cost-effectiveness, environmental sustainability, and availability from renewable resources. Outstanding functional properties, including enhanced complexation, chelation, occupancy, adsorption, and separation have been demonstrated by these recently developed biomaterials [120].

Functional groups embedded within hydrogel networks are key determinants of their chemical reactivity and capacity to adsorb heavy metal ions. Among these, nitrogen-containing groups such as amines play an important role; the lone pair of electrons on the nitrogen atom facilitates coordination with cationic metal ions. These groups are typically introduced through functionalization techniques such as formaldehyde treatment, gamma irradiation, or atom transfer radical polymerization. In addition to nitrogen-based functionalities, oxygen-containing groups, particularly hydroxyl groups, also contribute to adsorption behavior, primarily through hydrogen bonding and weak acid–base interactions. Though less reactive than other groups, hydroxyls enhance the overall hydrophilicity and polarity of the hydrogel matrix. Carboxyl groups, on the other hand, are widely recognized as the most effective sites for binding heavy metals. Under alkaline conditions, the carboxyl moiety loses a proton to form a negatively charged carboxylate ion (R-COO-), which exhibits strong affinity for divalent metal cations. Methods such as etherification, surface grafting, and TEMPO-mediated oxidation are often used to increase the density of carboxyl groups in hydrogel structures by converting primary hydroxyls into carboxyl functionalities [121].

An additional sulfur-containing group: an electronegative sulfur atom is singly bonded to the -OH group and double-bonded to two oxygen atoms, forming a sulfur-containing functional group called a sulfonic acid ($R\text{-}SO_3H$). A sulfonate group ($R\text{-}SO_3\text{-}$) is created when the hydrogen atom splits off from the sulfonic acid. The hydrogel's surface becomes negatively charged when a sulfonate group is covalently attached, regardless of the media pH. In one study, a hydrogel synthesized using 2-acrylamido-2-methylpropane sulfonic acid (AMPS) and exposed to $^{60}$Co gamma-ray irradiation demo2+trated strong adsorption affinity for $Co^{2+}$, $Mn^{2+}$, $Cu^{2+}$, and $Fe^{3+}$ ions [122].

## 6 CONCLUSION

Biopolymer-based hydrogels have demonstrated considerable potential as eco-friendly, efficient materials for removing heavy metals and dyes from wastewater. Their high adsorption capacity, structural tunability, and environmental safety make them attractive candidates for sustainable water treatment. This review highlighted the critical role of functional groups, such as carboxyl, amino, and hydroxyl, in enabling selective interactions with metal ions through mechanisms like ion exchange, chelation, and electrostatic attraction.

Despite substantial progress, several challenges remain unresolved. These include the limited mechanical stability of some hydrogel systems, reduced adsorption performance under complex water chemistries, and the lack of standard evaluation protocols for real-world applications. Moreover, most current studies are limited to lab-scale investigations and often lack assessments of regeneration or recyclability.

Future research should focus on developing hybrid hydrogels with enhanced structural integrity, optimizing crosslinking methods, and integrating biochar or nanomaterials to enhance performance. There is also a need for more field-scale studies, life cycle assessments, and techno-economic analyses to facilitate large-scale implementation. Overall, biopolymer-based hydrogels hold strong promise for advancing the next generation of green technologies for environmental remediation.

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### Conflict of interest

The authors declare no conflict of interest.

### Consent to publish

N/A

### Ethical approval

N/A

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