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Review article

Removal of arsenic (III) and (V) from water bodies by using biopolymers via adsorption: A review

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Abstract. Arsenic, an element found in the Earth's mantle, can be highly toxic, especially in its As (III) form. It enters our food chain through human activities like melting metals, using arsenic-based pesticides, and natural processes like volcanoes and rock breakdown. Consuming too much arsenic is extremely dangerous, impacting many countries worldwide. To tackle this issue, various methods like filtering, adding chemicals, and using electricity have been developed to clean arsenic-contaminated water. Among these, adsorption is a standout approach due to its simplicity and effectiveness. Biopolymers from living sources offer a natural solution, easily tweaked for arsenic removal. These biopolymers contain functionalities that can strongly latch onto toxic materials, acting like magnets. By customizing them with compounds like titanium dioxide (TiO_2), magnetite (Fe_3O_4), and others, they become even better at capturing arsenic, shaped into tiny particles or beads. This adaptation makes biopolymers a promising choice for cleaning arsenic from water. This review focuses on ways to clean water, specifically exploring how materials like chitosan, alginate, and modified cellulose can be used to remove arsenic by adsorption. It investigates how these materials work under different conditions, highlighting important details. By sharing these insights, this article contributes to the ongoing efforts to ensure cleaner water resources.

Keywords: *As (III), As (V), adsorption, biopolymers, heavy metal, wastewater treatment*

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1. Introduction

Arsenic is a group V element and exists as the 20th most commonly available element that naturally occurs in the oxidation states of +5, +3, and less frequently as 0 or –3 [1, 2]. Water supplies typically contain inorganic forms of arsenic, the most widespread of which are arsenite (AsO_3^{3-}) and arsenate (AsO_4^{3-}), also known as As (III) and As (V) [3]. Arsenite is approximately 10 times more harmful to plants and animals than arsenate. In aqueous solutions, arsenite is not particularly stable and readily oxidizes chemically using $KMnO_4$ or Cl_2, electrochemicaly and photochemically to form arsenate. Oxidation of arsenite to arsenate before the eradication procedure is required for the maximum elimination of arsenic from drinking water supplies because arsenate can be removed with greater efficiency than arsenite [4].

The typical amount of arsenic in the Earth's upper surface is approximately 5 mg/kg and is extensively distributed. Arsenic seeps into groundwater through biological processes, weathering of rocks, geochemical interactions, and volcanic emissions. Burning fossil fuels, melting metals, and the widespread use of arsenic-containing herbicides, pesticides, and insecticides are all anthropogenic sources of arsenic pollution [5]. More than 100 nations have detected arsenic pollution in groundwater [6]. After doing research and analyzing data from all around the world, The United States Environmental Protection Agency

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reduced the arsenic maximum contamination limit in potable water from 50 to 10 µg/l [7]. Up to 0.4 mg per day is the maximum tolerance limit for its assimilation. The ability of arsenic to interact with the sulphydryl groups of the enzyme system, which prevents the thiol groups of enzymes from executing their function, is one of the causes of arsenic toxicity [4]. Arsenic toxicity is a complex issue, categorized into acute and sub-acute toxicity. Acute poisoning necessitates immediate medical intervention, typically resulting from consuming contaminated food or water. At the same time, sub-acute toxicity affects various systems, including respiratory, gastrointestinal, cardiovascular, neurologic, and hemopoietin as shown in Figure 1 [8].

Various treatment methods have been tried for the eradication of arsenic from actual polluted water, including coagulation-flocculation, adsorption, filtration, and electrocoagulation [9–12]. These remediation methods, however, have their drawbacks, such as the creation of harmful by-products, the requirement for pretreatment, the need for an extra oxidation step to more effectively get rid of neutral arsenite ions, interference from other ions, extremely high operational costs, low efficiency, and lengthy duration [13]. Adsorption is a favorable practice designed for lowering the level of arsenic pollutants in the aqueous media owing to its wide adaptability, easy functioning, extraordinary efficacy, and sustainability. There have been many studies on the use of activated charcoal to reduce arsenic pollution. Contrarily, each gram of activated carbon adsorbs only a minor amount of metal ions [14–16]. They are complex to recover. Given that, employing activated carbon is expensive, which makes it difficult to use in developing countries [17, 18]. These problems can be addressed by implanting nanoparticles on biopolymers such as chitosan, sodium alginate, cellulose, lignin, pectin, and gelatin to get better results [19]. They act as good chelating agents due to the presence of functional groups like sulphonic, amine, carboxylic, phenolic, and hydroxyl groups. These biopolymers are preferred as sorbents due to their chemical stability, distinctive framework, physicochemical attributes, high sensitivity, and specificity as a consequence of the incorporation of chemically responsive groups. Furthermore, these substances are readily accessible, renewable, biodegradable, and able to chemically and physically bind an extensive range of molecules [20, 21].

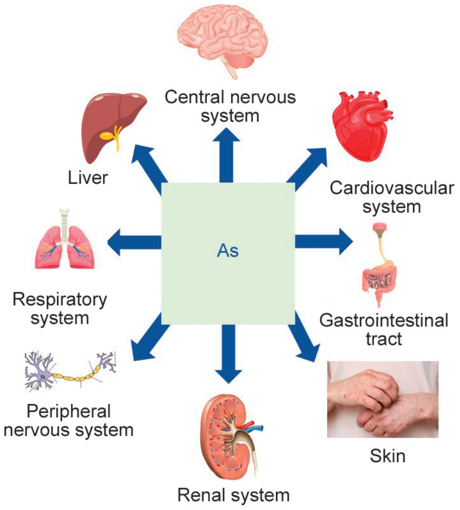


Figure 1. Effect of arsenic on different organs of the human body.

In this review, various methods such as adsorption, coagulation and flocculation, membrane filtration, and electrochemical are discussed to remove the arsenic from wastewater. Along with the synthesis of adsorbents by modifications in biopolymers like chitosan, alginate, and cellulose are discussed, and their maximum adsorption capabilities at different conditions are elaborated.

2. Methods for removal of arsenic from water

2.1. Coagulation and flocculation

Coagulation is an effective method for eliminating arsenic from soil and seawater [22]. Substantial wastewater preprocessing and using unprocessed chemicals are not required in this technique. For such treatments, pre-oxidation and pH adjustment methods are required. This arsenic extraction method can be made simpler if the water characteristics allow it by skipping the sedimentation phase [23]. In this method, arsenic that has been dissolved is converted to globules by adding positively charged coagulants that neutralize the negative charge of colloidal particles. Colloids thus build up into larger particles that precipitate and can be separated by filtering. By removing several additional suspended pollutants and toxic substances in addition to arsenic, this technique improves the quality of water [24–27]. Water contaminated with arsenic produces water-unsolvable artificial oxides due to the dissolution of other flocculants. These oxides should co-precipitate with arsenic for effective cleaning, resulting in gelatinous deposition and removal of water-unsolvable forms [28].

Arsenic removal techniques that involve chemical coagulant agents like alum and ferric chloride are proven to be the most beneficial methods [29]. While using $FeCl_3$ as a coagulant, the occurrence of organic matter reduces the arsenate extraction at pH 8 and 9. Alum has a smaller pH range than ferric chloride for eliminating arsenic [30]. Details on the various coagulants used to remove arsenic are included in Table 1, along with their % removal rates. It is easy and efficient to separate charged particles from the liquid using coagulation-flocculation, as shown in Figure 2.

2.2. Membrane filtration

Membranes have certain morphological traits because of their structure and composition, including pore diameter, pore structure, homogeneity, hardness, thickness, penetrability, and selectivity. The following qualities should also be present in them such as mechanical and chemical resistance, greater penetration, specificity, extended durability, and inexpensive cost. [33]. Various techniques have been used for arsenic removal by using membrane systems such as filtration and electrical repulsion. Microfiltration and ultrafiltration may not be able to immediately remove arsenic species because they have smaller pore sizes. Arsenite and arsenate species can be removed using nanofiltration (NF) because of size omission. Nanofiltration is commonly employed to remove divalent samples [11, 34, 35]. Arsenic removal from groundwater was studied by using a loose nanofiltration membrane. Due to the dominance of Donnan exclusion over steric exclusion in regulating the arsenic removal capability of the membrane, arsenate was rejected at a significantly higher rate than arsenite [36]. An inexpensive kaolin was used to create the porcelain hollow fiber membrane. As a consequence, at an input temperature of 60 °C, a significant permeate flow with complete arsenic rejection was generated, satisfying the essential requirement of a maximal pollution level of 10 ppb. Additionally, levels of arsenic up to 1000 ppm and various arsenic pH readings were looked at, but no arsenic was found [37]. Table 2 describes the various membranes used to remove different kinds of arsenic from wastewater.

2.3. Electrochemical methods

Arsenic removal can be accomplished by using electrocoagulation. This electrochemical technique is believed to achieve a significant reduction in the quantity of solid sludge generated as compared to the complete eradication of arsenic since electrocoagulation delivers a relatively small volume of ferrous ions. The electrocoagulation process is shown in Figure 3. However, throughout the electrocoagulation process, the creation of ferric ions may be efficiently controlled by altering the operating parameters [42]. Combining electrodes like aluminum and iron in

Table 1. Percentage capacity of several coagulants.

The type of arsenic removed	Coagulant	Initial concentration [mg/l]	Coagulant dosage [mg/l]	pH	Percentage removal [%]	Reference
As (III)	$FeCl_3$ chemical coagulation	0.1	1.5	6.5	98	[9]
As (V)	$FeCl_3$ chemical coagulation	0.1	1.5	6.5	70	[9]
As (V)	Aluminum sulfate	0.5	42.0	7.0	99	[31]
As (III)	$FeCl_3$	1.0	30.0	7.0	60	[32]
As (V)	$FeCl_3$	1.0	30 .0	7.0	90	[32]
As (III)	Chitosan with $FeCl_3$	1.0	0.5	7.0	80	[32]
As (V)	Chitosan with $FeCl_3$	1.0	0.5	7.0	100	[32]

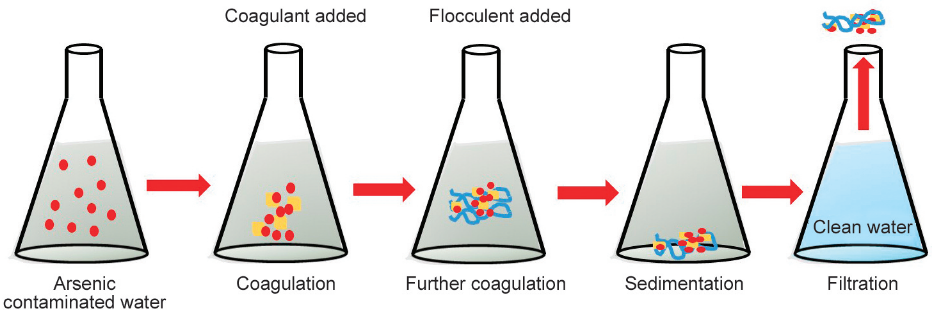


Figure 2. Graphical illustration of arsenic removal using the process of coagulation and flocculation.

Table 2. Various membrane-based separation techniques for arsenic removal.

Kinds of arsenic removed	Membrane-based separation technique	Initial concentration	Membrane type	pH	Removal capacity [%]	Reference
As (III)	Nanofiltration	118 ppb	Thin filmed composite (TFC) with spiral coiled and aromatic polyamide	8	94.07	[38]
As (III)	Nanofiltration	100 ppb	Flat sheet thin film NF	7.5	67.72	[39]
As (III)	Reverse osmosis	100 ppb	RO membranes	7.5	97–99	[39]
As (V)	Cross-flow nanofiltration	150 ppb	Polyamide thin filmed composite membrane	7	97–100	[40]
As (V)	Nanofiltration	120 ppb	Thin filmed composite with spiral coiled and aromatic polyamide	8	98.35	[38]
As (V)	Nanofiltration	6 mg/l	Commercial thin film composite (TFC) NF-300	8	99.8	[41]

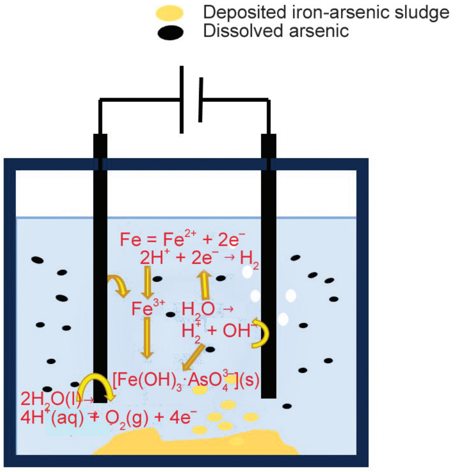


Figure 3. Schematic representation of arsenic removal using iron via electrocoagulation.

one electrochemical system is another method for electrocoagulation to remove arsenic from groundwater [43]. For the extraction of As (III) and As (V), electrocoagulation generates ferromagnetic and amorphous iron hydroxide pieces. In the laboratory, an electrocoagulation reactor removes 99% arsenic in 90 s or less [12].

A technique called continuous electro-deionization employs electric potential to remove charged substances from water or any other liquid. It combines electro-dialysis with ion exchange isolation [44]. Using a set potential difference, the mechanism for arsenic removal as a function of the state of the system was studied. Potential difference varying from 0.8 to 5.2 V was introduced, having a phase variation of 0.2 V and a 3 h perseverance duration to investigate the cell saturation frequency. To develop an ion transport and ion econometric approach, each chamber of the continuous electro deionization shell was evaluated for conductance, ionic strength, and pH at every conceivable step [45–47]. The ion exchange or electro-dialysis technique is being done in various steps by circulating arsenic solution via the central cabin as well as the two neighboring chambers. OH^- ions of resins substitute arsenate ions, which are further produced again due to the splitting of water at the membrane-solvent interface. It depends on changes in conductivity, pH, and concentration of arsenic in each chamber [48].

2.4. Adsorption

Adsorption is an economical and efficient method of eliminating arsenic, particularly in underdeveloped nations that struggle with a lack of trained labor and power production. Its efficiency can reach up to 95%. The pH, temperature, and initial concentrations of adsorbent and adsorbate are some of the factors that impact the productivity of the adsorption process. [49].

Various adsorbents have been synthesized for arsenic eradication from water are shown in Table 3. TiO_2 and TiO_2-based products have been used to remove both organic and inorganic arsenic. Currently, it seems that titanium dioxide-based arsenic extraction techniques focus on the oxidation of arsenite (As (III)) to arsenate (As (V)) in the presence of photocatalysts [50]. Innovative synthetic materials, compared to conventional adsorbents, are better alternatives for the efficient removal of arsenic, as evidenced by enhanced recycling and reuse and greater partition coefficient. Organometallic complexes, graphene oxide, carbon nanotubes, and other forms are examples of these materials. Innovative methods were thus found to hold great effect as potential substitutes for several conventional substances for the effective recovery of arsenic from drinking water or wastewater [51, 52].

Table 3. Various adsorbents and their adsorption capacities for arsenic removal.

Adsorbate	Adsorbent	Adsorbate concentration [mg/l]	Adsorbent dosage [g/l]	Time [min]	Removal [%]/ Adsorption capacity [mg/g]	Reference
As (III)	Zirconium (IV) oxide-ethanolamine	10–100	0.0007	50	98%	[53]
As (III)	Magnetic γ-Fe_2O_3 nanoparticles	10	1.6	30	74.83 mg/g	[54]
As (III)	Perilla leaf-derived biochar at 300 °C	6.0	1.0	120	4.71 mg/g	[55]
As (III)	Perilla leaf-derived biochar at 700 °C	6.0	1.0	120	11.01 mg/g	[55]
As (V)	Mesoporous alumina	44.703	0.02	120	36.6 mg/g	[56]
As (V)	Leonardite	5	0.02	180	8.40 mg/g	[57]
As (V)	Fe/Cu nanoparticles	1–5	0.1	30	21.32 mg/g	[10]

2.5. Biosorption

Biosorption is the process of using biological resources or the materials derived from them, such as various agricultural waste products, to remove pollutants from water, including metals and metalloids. Based on the qualities of the biomass, the physicochemical process of biosorption may result in a variety of physical bindings (such as electrostatic interactions, hydrogen bonds, and van der Waals forces) as well as chemical bindings (such as ionic and covalent bonds) [58, 59]. Generally, the benefits of improved biosorption include extraordinary efficacy for low metal/metalloid concentrations, easy way of operating, capability, high specificity, the potential for metal/metalloid retrieval, high availability, and applicability in a broad spectrum of experimental environments [60, 61]. Several disadvantages of untreated biomasses make them unsuitable for conventional process equipment such as high sensitivity, poor solubility, instability, release of colors in treated water, and low sorption [62–64]. Untreated biomasses need modification to get over these drawbacks for commercial and industrial uses. Some natural adsorbents that have been used to extract from water are included in Table 4.

Various methods for arsenic removal include coagulation/precipitation, adsorption, ion exchange, and membrane filtration. Comparison of these methods for and their advantages and disadvantages is shown in Table 5.

3. Biopolymers and their modifications

Biopolymers, also known as natural polymers, are defined as polymers created by living things during their normal growth cycles. They are created inside cells through intricate metabolic processes [74]. These macromolecules have an incredibly diverse spectrum of uses, from cellular information storage

Table 4. Several biosorbents with their maximum adsorption capacities.

Type of arsenic species removed	Bio sorbent used	Conditions for experiment	Adsorption capacity	Reference
As (III)	Cellulose in the calcium/aluminum layered double hydroxide	pH: 6 Temperature: 25 °C Bioadsorbent used: 2 g/l	10.9 mg/g	[65]
As (III)	Cellulose in the zinc/aluminum layered double	pH: 6 Temperature: 25 °C Bioadsorbent used: 2 g/l	30.2 mg/g	[65]
As (V)	Cellulose in the calcium/aluminum layered double hydroxide	pH: 6 Temperature: 25 °C Bioadsorbent used: 2 g/l	42 mg/g	[65]
As (III)	Biomass of *Bacillus cereus*	pH: 7.5 Temperature: 30 °C Bioadsorbent used: 6 g/l	85%	[66]
As (V)	Organic orange peel	pH: 6.5 Temperature: 20 °C Bioadsorbent used: 4 g/l	68%	[67]
As (V)	Charred orange peel	pH: 6.5 Temperature: 20 °C Bioadsorbent used: 4 g/l	98%	[67]

Table 5. Comparison of different methods for removal of arsenic.

Method	Advantage	Disadvantage	Reference
Coagulation and flocculation	Affordable start-up cost Easy manufacture of raw materials No need for constant monitoring	Complex procedure Disposal of toxic sludge Pre-oxidation can be needed Highly skilled workers are required	[68]
Membrane filtration	No solid wastage No use of chemicals Greater efficiency Removes other contaminants, too	High investment required Consistent care is needed Water quality needs to be re-adjusted regularly Membrane fouling	[11, 68]
Electrochemical methods	Less space required Sustainable method Easy to function	Expensive setup Greater sludge formation Greater consumption of energy	[11, 69]
Adsorption	Effective removal of As Inexpensive Easy to perform Possibility of regeneration	Adsorbents need to be replaced after 4 cycles pH and co-existing ions affect the adsorption capability Pretreatments can be required	[70, 71]
Biosorption	High reusability Low working cost High efficiency	Loss of mass after recycling Factors like pH and the presence of other heavy metals affect performance Depend on species of As present in water	[72, 73]

to protective shells [75]. Alginate, chitosan, cellulose, and other natural polymers have been studied in recent research, showing important benefits because of their strong affinity and adsorption ability for different contaminants. The novel composites show enhanced mechanical strength, durability, and hydraulic qualities after incorporating biopolymers as support [76]. Table 6 gives a brief description of biopolymers, which are further discussed as a modified composite in this literature review [75].

3.1. Chitosan based systems

Chitosan is a natural polymer made up of chitin, which is the second most abundantly found polymer in the world. The majority of its structure as shown in Figure 4 is composed of 1,4-linked glucosamine and *N*-acetylglucosamine residues. It can be synthesized by deacetylating chitin, which is produced from the shells of insects, crustaceans, and other sources [77]. It has a flexible polymer chain, is very hydrophilic, and has several hydroxyl and amino groups that could be used as adsorption sites [78]. Its abundance in nature, low processing requirements, and being a by-product of the fishing industry make it an economical sorbent [79]. Chitosan has gone through several modifications to increase its acid stability and/or ability to bind arsenic [80]. In Table 7, some of the effective chitosan-based composites are discussed for the removal of arsenic from groundwater.

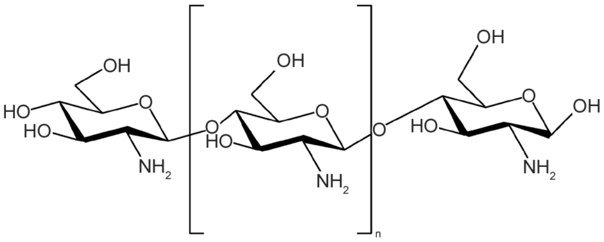
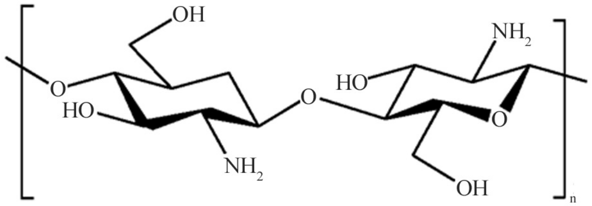
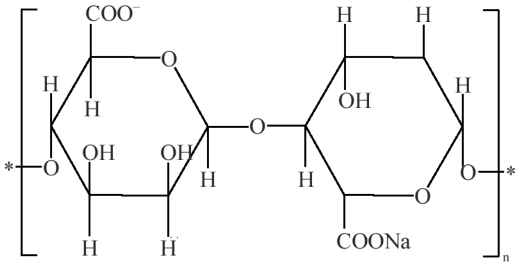
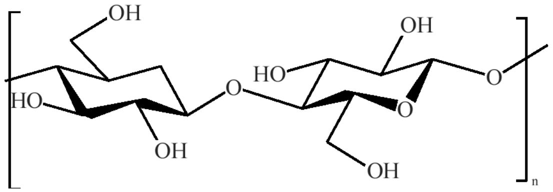


Figure 4. Structure of chitosan.

3.1.1. Iron-chitosan composites

Chitosan with iron (Fe) has been used in several forms, such as granules, flakes [81], nanoparticles (NPs) [82], and microspheres [83] for arsenic remediation. Iron-chitosan flakes and granules were tested for inorganic arsenic removal through adsorption at pH 7.0 [81]. The results showed that the monolayer adsorption capacity of iron-chitosan flakes is greater than the capacity of iron-chitosan granules. The adsorbent was also effectively used to remove total inorganic arsenic from an actual groundwater sample down to 10 mg/l. Iron manganese (Fe-Mn) binary oxide was impregnated into the matrix of chitosan to create beads. The results showed that a large amount of arsenic was removed due to adsorption onto the Fe-Mn oxide component [84]. Recent research shows the use of magnetic chitosan composites prepared by using iron for the adsorption of arsenic [85]. By using electrospraying technology, magnetite/ferric hydroxide microspheres were created as new and cost-effective arsenic adsorbents. Adsorption was very quick, and results indicated that the microspheres had superparamagnetic properties and microporous structure, giving them a high As (III) adsorbing capacity and a high efficiency for

Table 6. A brief description of biopolymers.

Name of biopolymer	Category	Primary source	Structure	Key properties
Chitosan (chitin)	Basic polysaccharides	Chitin from arthropod shells is deacetylated by a base		After cellulose, the most prevalent carbohydrate on Earth – Chitin has a low degree of solubility (such as ionic liquids, dimethylacetamide (DMAC)/LiCl, $CaCl_2$/methanol) – Degree of deacetylation affects properties of chitosan – Several functionalities are present
Alginate	Acidic polysaccharide	brown algae are extracted using hot alkali		– Acts as a copolymer for guluronate and mannuronate – Shows metal cations cross-linkage – Insoluble in water, alginic acid is soluble in ions such as Na^+, K^+, and NH_4
Cellulose	Neutral polysaccharides	structural biopolymer in plants, also produced by some bacteria		– Linear polymer, the most abundant naturally occurring polymer-insoluble in many solvents, including water

separation [83]. A few more studies have been done in this area to evaluate magnetic biopolymer adsorbents for arsenic [86, 87].

3.1.2. Alumina-TiO_2-chitosan composites

TiO_2-impregnated chitosan beads (TICB), a new sorbent for arsenic, have been created and effectively tested [88]. TICB eliminated 2198 µg As(III) and 2050 µg As(V) without using UV rays. When exposed to UV rays, TICB was capable of photo-oxidizing arsenite to arsenate, a less hazardous form of metal that is also easier to sequester. Additionally, UV light led to improved arsenic elimination. Metal oxide-impregnated chitosan beads (MICB) comprising metal-oxides like nanocrystalline alumina and titanium dioxide were examined [89]. This adsorbent used the photocatalytic activity of TiO_2 to oxidize arsenite to arsenate and the high adsorption capacity of Al_2O_3 for arsenate to produce a removal capacity that is greater than either metal oxide alone. While preserving a desirable life cycle, MICB was discovered to be a potent and more effective eradicating agent for arsenic than previously reported.

3.1.3. Molybdate-chitosan composites

Molybdate-treated chitosan beads were demonstrated to be efficient, even at low residual arsenic concentrations with high relative sorption capabilities [90]. The experimental results for As(V) sorption on molybdate chitosan composite were explained at pH 3 by using Langmuir adsorption isotherm. In the pH range of 2 to 4, arsenate ions were strongly adsorbed with little molybdate ion release. Another finding on molybdate-impregnated chitosan beads (MICB) showed that prepared composite promoted the adsorption of both As(V) and As(III) at pH 5 [91]. With adsorption capabilities of 1.98 and 2.00 mg/g, respectively, it effectively removed arsenite and arsenate.

3.1.4. Volcanic rocks and chitosan composites

Rocks from volcanoes have been used with chitosan for effective remediation of arsenic from groundwater. Chitosan and volcanic rocks in three diverse ratios – 1:2, 1:5, and 1:10 are mixed, and their adsorption capability of arsenic has been examined. The effective mixing proportion for chitosan and volcanic rocks was 1:5, with the highest adsorption capacities of 0.72 mg/g for Ch-Rs (Chitosan–red scoria) and 0.71 mg/g for Ch-Pu (Chitosan–pumice). Both blends could eliminate 93% of As(V) solution in a broad range of initial pH standards, removing the requirement for pH regulation in practical applications. The results suggested that such combinations must be researched more as a viable long-term strategy for eliminating arsenate from water [80]. In Table 7, some of the effective chitosan-based compounds are discussed for the removal of arsenic from groundwater.

3.1.5. Chitosan ion-imprinted composites

Molecular imprinting technology is a rising technique in which template molecules use reversible immobilization to form sites where recognition of adsorbate occurs on macromolecular polymers. Beads were synthesized by using α-Fe_2O_3 and chitosan having As(III) imprinted on them. According to the batch test results, the maximum adsorption capacity was found to be 6.18 mg/g at pH 5, and the Langmuir equation evaluated a maximum adsorption capability of 9.355 mg/g. It was deduced that the prepared beads preferred arsenite for adsorption, which was removed more than non-imprinted chitosan composites [92]. In another study, arsenite imprinted composite nanoparticles were created in the presence of magnetite, *N*-(2-hydroxyl)propyl-3-trimethyl ammonium chitosan chloride using the micro-emulsion method [93]. According to Sips isotherm, the prepared nanoparticles exhibited a maximum adsorption capacity of 11.52 mg/g at pH 6 and remarkable regeneration ability. Even after 10 recycles, their adsorption capability was more than 75%. Overall, nanoparticles demonstrated good adsorption ability in a broad range of pH and were selective towards arsenite in the presence of other ions. A chitosan resin was prepared to remove As(III) from Laminaria japonica Aresch juice [94]. The resin efficiently removed arsenic and other ions except Zn^{2+}. In 4 h, equilibrium was attained, and the minimum adsorption was up to 4.16 mg/g at pH 6. The Fourier transform infrared spectroscopy (FT-IR) spectra showed that amino and hydroxyl groups on chitosan helped as coordinating sites for adsorption. Only 13% of adsorption ability was lost after 10 regeneration cycles. The prepared resin was found to be a promising adsorbent for arsenic removal. As ion imprinting technique is a new technique, it needs more exploration in the area of chitosan-based arsenic adsorbents.

3.1.6. Chitosan-based hydrogels

Hydrogels are three-dimensional structures that have the ability to absorb large quantities of water due to their polar nature. Recently, hydrogels prepared with biopolymers are getting a lot of attention for water remediation due to their functionalities [95]. Chitosan hydrogels were prepared with grafted *N*-vinylcaprolactam/*N*-*N*-dimethylacrylamide for the adsorption of arsenic and fluoride. At neutral pH conditions, arsenic was removed between 40% to 45% with a maximum capacity of 0.0022 mg/g due to electrostatic attractions between negative ions and the hydrogel surface. It was concluded that results can be improved by working on factors like concentration of adsorbate and adsorbent, pH, and contact time [96]. Another hydrogel of chitosan incorporating furfuraldehyde was synthesized and examined as a low-cost adsorbent for arsenate removal. At optimum conditions, the initial adsorption rate was found to be 0.1479 g/(mg·min). The hydrogel selectively removed As (V) in the presence of other ions like Ni (II), Cr (III), Cd (II), Pb (II), and As (V). The results show that prepared hydrogel can act as an environment-friendly, inexpensive, fast, and better option for arsenic removal from groundwater [97]. Light-cured methylated chitosan and gelation hydrogels were evaluated for arsenic and lead removal. The results suggested that hydrogels prepared with renewable substances are a good option to be used for heavy metal sorption as they exhibited high swelling properties and adsorption capacity [98]. Magnetic chitosan hydrogel beads also have been prepared and tested for arsenate removal. The results indicated the highest adsorption capability of 66.9 mg/g at neutral pH [99]. Overall, chitosan hydrogels provide excellent results for arsenic removal from aqueous media [19]. Chitosan can be incorporated easily by using simple procedures and forming cost-effective adsorbents. However, there are several drawbacks to using chitosan, such as it is less selective as well as less soluble in acidic environments, and has low mechanical strength [100]. After being modified with different material show remarkable adsorption capability, such as the combination of chitosan with volcanic rocks demonstrates better stability and adsorption efficiency in a broad range of pH [80]. Figure 5 demonstrates the adsorption mechanism of inorganic species of arsenic using chitosan from contaminated water.

3.2. Alginate-based systems

Alginate is a readily available polymer having an anionic nature, commonly derived from brown seaweed [107]. It forms by co-polymerization of α-L-guluronic and β-D-mannuronic acid residues arranged in various ratios and sequences [108]. When combined with divalent ions like calcium, alginate forms gels, and the amount and size of blocks of adjacent L-guluronic acid residues (G-blocks) in the polymeric chains substantially correspond with the gel-forming capabilities [109]. Making these gel beads is as simple as letting sodium-alginate solution

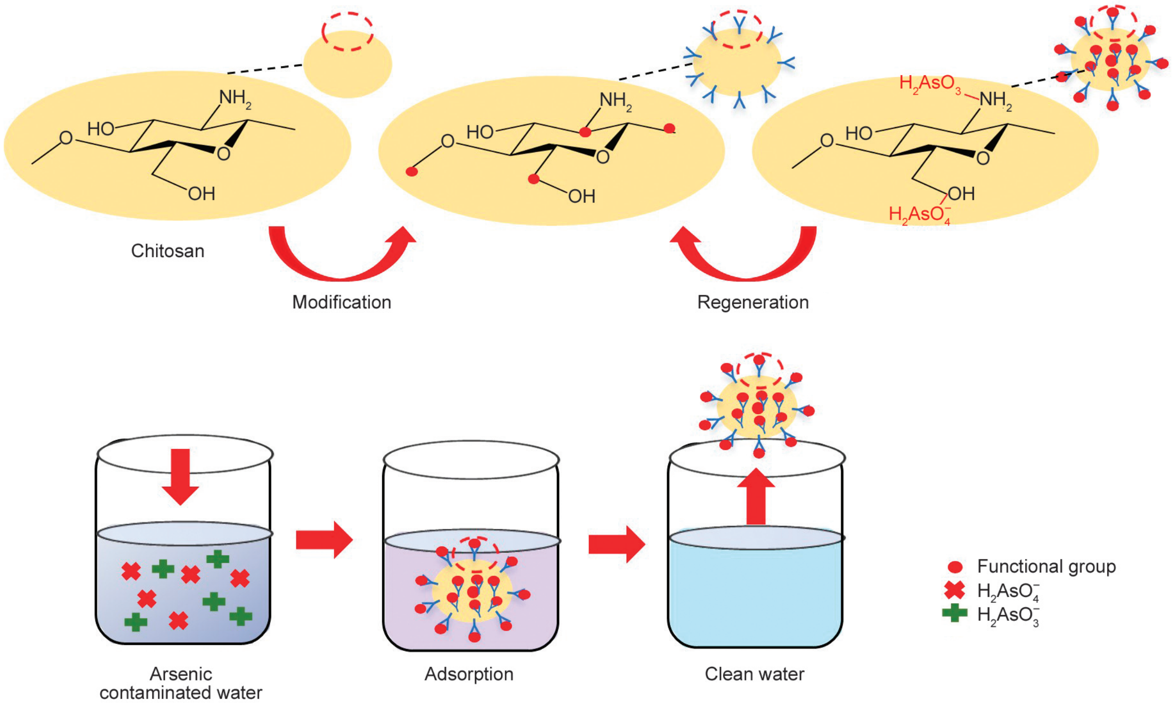


Figure 5. Mechanism of adsorption of arsenic on chitosan.

Table 7. Modified chitosan composites and their maximum adsorption capacities.

Type of compounds	Method of preparation of composite	pH	Maximum sorption capability of As (III) [mg/g]	Maximum sorption capability of As (V) [mg/g]	References
Chitosan sorbent	Chitosan beads were prepared using the precipitation method.	5	1.94	1.83	[101]
Iron-chitosan flakes	The mixture of $Fe(NO_3)_3$, NaOH, and chitosan flakes were prepared. The mixture was stirred using a magnetic stirrer, and N_2 gas was purged.	7	16.15±0.32	22.47±0.56	[81]
Iron-chitosan granules/beads	Beads were prepared using the precipitation method.	7	2.32±0.05	2.24±0.04	[81]
TiO_2-impregnated chitosan beads	TiO_2 nanopowder was added to the chitosan solution. Beads were prepared by loading this solution in a syringe and dispensing them in NaOH solution.	6.61 (for As (III)) 7.02 (for As (V))	6.2	4.92	[88]
Alumina-chitosan grafted polyacrylamide nanoparticles	Alumina nanoparticles were prepared by reverse microemulsion, and in situ dispersion was used to immobilize them on a chitosan-grafted polyacrylamide matrix.	7.2	–	6.65	[102]
Chitosan zerovalent iron nanoparticle (CIN)	Nanoparticles were prepared by reduction of Fe in the presence of $NaBH_4$ using chitosan as a stabilizer.	7	94±1.5	119±2.6	[103]
Magnetic-chitosan-ferric hydroxide microspheres	Fe_2O_3 NPs were prepared by co-precipitation. Chitosan, $FeCl_3 \cdot 6H_2O$, and Fe_3O_4 mixture was electro-sprayed on the nanoparticles.	7.0±0.1	8.47	–	[83]
Magnetite-chitosan beads	Fe_3O_4 NPs were prepared by using the solvothermal method. The template method was used to prepare magnetic chitosan beads.	6.8	35.3	35.7	[82]
Chitosan-clay-magnetite composite	Nano-magnetite (NMT) and Heulandite (HE) were mixed with chitosan solution and then dropwise poured into the sodium tripolyphosphate (STPP) solution.	3–9	–	5.9	[104]
Fe-Mn binary oxide impregnated chitosan beads	Powdered Fe-Mn oxide was used to prepare nanoparticles, and then fabrication was done in the chitosan mixture.	7.0±0.1	54.2 (batch test) 1.76 (column test)	39.1 (batch test) 0.82 (column test)	[84]
Molybdate-impregnated chitosan beads	Chitosan beads were prepared by precipitation method and then were brought in contact with ammonium heptamolybdate.	2–3	70	230	[105]
Chitosan- graphene oxide composite	Graphene oxide was prepared from graphite. Stock solutions of arsenic were diluted to prepare the required sample solution.	4.3–6.5	64.2	71.9	[106]

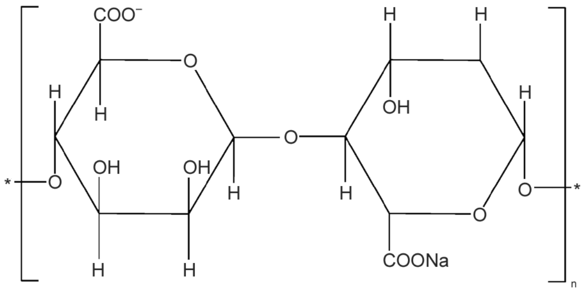


Figure 6. Structure of sodium alginate.

droplets fall in a calcium chloride bath [108]. The structure of sodium alginate is shown in Figure 6. Metals, including Pb^{2+}, Zn^{2+}, Cd^{2+}, and Ni^{2+}, have been eliminated from water by adsorbing them on alginate-based adsorbents [110]. This literature reviews the alginate-containing adsorbents for arsenic elimination.

3.2.1. Calcium alginate beads

Beads prepared using Ca-alginate were studied to eliminate arsenic from diluted aqueous solutions [111]. As (V) ion adsorption followed the Langmuir isotherm model with an adsorption coefficient of 22.64 mg/l. The adsorption of arsenate was quite sensitive to the pH of the solution, and it indicated its maximum absorption value at pH 6.0. Three distinct adsorbent materials, including Ca-alginate beads (G), KOH-activated carbon-based apricot stones (C), and Ca-alginate-activated carbon composite beads (GC), were examined for the eradication of arsenic. The thermodynamic variables and the observation revealed that a pseudo-second-order process was used in the adsorption of arsenic ions. C showed the highest degree of porosity, total pore volume, and surface area. Meanwhile, G had the property of highly acidic functional groups on it. The maximum As (V) adsorption was observed in GC, indicating its efficiency as an adsorbent over Ca-alginate beads and KOH-activated carbon-based apricot stones [112].

3.2.2. Calcium alginate magnetic composites

A magnetic sorbent was prepared using alginate and was investigated for arsenic elimination. The sorbent was prepared by encapsulating calcium alginate around magnetite Fe_3O_4. According to X-ray photoelectron spectroscopy (XPS) analysis, absorption on the sorbent was followed by the reduction of arsenate into arsenite. According to these results, the OH^- part of the Ca-alginate and the oxygen in the lattice of magnetite both played significant parts in the sorption of arsenate ions onto the sorbent. Scanning electron microscopy (SEM) analysis showed that the sorbent surface had numerous protuberances and pores, which facilitated the adsorption and diffusion of As (V) ions onto the composite. All the analyses justified the function of calcium alginate magnetic beads as a sorbent for arsenate in an aqueous medium [113].

3.2.3. Ca-alginate-nanoscale zero-valent iron (NZVI) composite

Ca-alginate with zerovalent iron having nanoscale dimensions composites was studied as a remarkable arsenic eradication material. In batch experiments, arsenic removal of 85 to 100% was accomplished in 2 h with initial arsenate values of 1 to 10 mg/l. According to the FT-IR study, arsenic was solely bound to iron and not to Ca-alginate. Studies with low arsenic concentrations in the range of 50–100 µg/l showed that prepared sorbent could successfully eliminate actual groundwater arsenic to levels below instrument detection limits [114].

3.2.4. Hydrous iron oxide (HIO) alginate composite

Alginate beads impregnated with hydrous Fe-oxide were used to examine inorganic arsenic elimination [115]. Amorphous hydrous iron oxide, prepared by the chemical precipitation method, was infused with alginate gels. At pH 6 to 9, arsenite removal by getting adsorbed onto hydrous Fe-oxide-alginate beads was most effective, but arsenate elimination was higher in acidic than in alkaline solutions. Both arsenite and arsenate adsorption efficiencies were largely enhanced with higher Fe loading. However, because of the reduction in bead surface area, the efficiency of arsenate adsorption at high loading was somewhat diminished. It can be deduced from the results that HIO-alginate beads can act as an effective adsorbent for eliminating arsenic.

3.2.5. Zirconium-oxide alginate beads (ZOAB)

Zirconia entrapped in alginate beads was examined for effective removal of both arsenic species and copper simultaneously. The maximal adsorption capacity of the prepared composite was 32.3, 28.5, and 69.9 mg/g for arsenite, arsenate, and copper, respectively. The presence of copper ions raised the positive charge on the composite surface, making arsenate sorption more successful than arsenite sorption. In conclusion, the produced composite was very effective in removing both anions and cations from the aqueous medium at the same time. ZOAB was considered to be able to handle a variety of contaminated waters, including cations and anions-contaminated groundwater, industrial wastewater, and acid mine drainage [116].

3.2.6. Alginate-clay composites

Sodium alginate beads constituting iron-modified sericite clay were prepared. Batch testing of the composite beads revealed significant As(V) absorption over a wide-ranging pH, and the highest amount of As(V) adsorbed was 5.780 mg/g. The sorption kinetic data showed that intra-particle diffusion was essential to the sorption process and that the sorption equilibrium must be reached after at least 12 h of contact time. Additionally, a fixed-bed column trial showed that iron-sericite beads could eliminate As(V) up to an acceptable standard under dynamic settings [117]. Another group tried iron-modified sericite clay with sodium alginate for the removal of arsenic and lead. The results showed that adsorption efficiency decreased with increasing pH. The composite exhibited maximum arsenic removal due to the presence of $Pb(OH)_2$ precipitates in the binary system [118].

3.2.7. Alginate ion-imprinted composite

Chitosan and alginate consisting of iron oxyhydroxide nano-adsorbents were prepared in a modified way [119]. For arsenite, alginate beads were found effective, and for arsenate, chitosan beads showed maximum adsorption. The results concluded that hydroxy and amino groups helped in adsorption, and only 2 g/l alginate beads were enough for water treatment. The prepared composite was found to be useful after several recycling procedures. In comparison, with two different commercially available adsorbents that have high adsorption capacities, the prepared beads exhibited arsenic adsorption capability higher than them. This showed the potential of chitosan and alginate ion-imprinted composites as valuable adsorbents for the removal of heavy metals from aqueous solutions.

3.2.8. Alginate-based hydrogels

Alginate hydrogel beads containing polyvinyl alcohol (PVA) and bentonite were prepared, having different concentrations of PVA and alginate. The results showed bentonite played an important role in arsenic adsorption. At short treatment times, beads with bentonite and a high concentration of alginate exhibited better results. At longer treatment times, both bead formulations containing bentonite displayed complete removal of arsenic. The results deduced that the prepared beads can act as a potential adsorbent for arsenic [120]. The ternary composite containing graphene oxide, sodium alginate, and yttrium was prepared and examined for adsorption of tetracycline and arsenic. For arsenate, maximum adsorption was found to be 273.39 mg/g due to ion exchange between OH^- and $H_2AsO_4^{2-}$ groups and hydrogen bonds formed with O-containing groups on the prepared composite. It was observed that the presence of tetracycline affected the adsorption of As(V) in the binary system. Overall, properties like reusability, easy separation, and adsorption efficiency can contribute to making it an excellent material for adsorption [15]. Akageneite nanorods enclosed in alginate beads were evaluated for arsenic adsorption, and the results suggested the prepared composite to be a fast and easy material for arsenic extraction from water in developing countries. Arsenite removal efficiency was found to be less than arsenate because of the weaker interaction of As(III) with akageneite. Whereas the mechanism of As removal and reusability of the hydrogel was not discussed [121]. A few more studies have been done on sodium alginate-containing hydrogels which are found to be effective for heavy metal removal from water [122–125].

Alginate adsorbents are found to be more effective towards arsenate because arsenite requires pre-oxidation for conversion into arsenate [126]. These adsorbents and their adsorbing capacities are shown in Table 8. Several composites work best in neutral pH; they cannot provide maximum adsorption in highly acidic or alkaline conditions [115, 127]. Moreover, reasons like the complexity of the procedure, running cost, reusability of adsorbents, and production of waste containing arsenic hinder its application on the industrial level [128]. In the case of ion-imprinted biopolymers, alginate should be explored in the area of arsenic removal.

3.3. Cellulose-based system

In nature, cellulose as shown in Figure 7 is the most copious biomass available, and it produces various adsorbents in both indigenous and derivative forms. Several vinyl monomers such as acrylamide, acrylic

Table 8. Alginate adsorbents and their adsorption capacities.

Type of compounds	Method of preparation of composite	pH	Maximum Sorption capability of As (III) [mg/g]	Maximum Sorption capability of As (V) [mg/g]	Reference
Water treatment residuals (WTRs) alginate beads	After chemical and thermal pretreatments of WTRs , they were mixed with an alginate solution and $CaCl_2$ was added for gelation.	3.0–7.0	3.4	2.9	[129]
Ca-Fe alginate beads	The alginic acid solution was added into $CaCl_2$ and $FeCl_3$ solution and agitated magnetically at 8.0±0.1 pH. After 24 h, beads were recovered and dried.	As (III) 4.0–8.0 As (V) 2.0	0.117	0.364	[130]
MnO_2 alginate beads	MnO_2 was obtained from a lithium-ion battery after its washing and drying, mixed with sodium alginate solution. Added this solution dropwise to $CaCl_2$ to get composite beads	6.5	2.0	–	[131]
Alginate- drainage sludge beads (AMDS)	Dispersion of goethite in alginate solution keeping the ratio of AMDS to alginate 1:9 (w/w). $CaCl_2$ was added to get the composite beads	2.0–9.0	18.25	21.79	[132]
Alginate and carboxymethyl cellulose beads	Carboxymethyl cellulose and sodium alginate were added to the $CaCl_2$ solution. After the formation of beads, they were washed and dried.	6.0	–	4.39	[133]

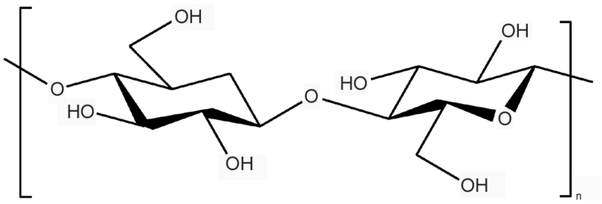


Figure 7. Structure of cellulose.

acid, acrylonitrile, *N*-isopropylacrylamide (NIPAM), and hydroxyethyl methacrylate can be grafted on the backbone of cellulose due to the presence of hydroxyl group [134]. These hydroxyl ions cause complex structures having crystalline and non-crystalline regions; the firmness of molecular chains and strong hydrogen bond interactions make cellulose difficult to melt and dissolve. Therefore, researchers, through chemical modifications, use cellulose and form new composites with applications in various fields [135]. Cellulose nanomaterials can easily replace petroleum-based adsorbents due to their properties like biodegradability, inexpensiveness, recyclability, and abundance [136].

3.3.1. Cellulose magnetic composites

Cellulose magnetic composites were prepared in a step method using Fe_2O_3, while cellulose was dissolved in urea, thiourea, and NaOH solution. The maximum adsorption capacity for arsenite was 23.16 mg/g, and arsenate was adsorbed up to 32.11 mg/g. At pH 6, arsenite showed the highest adsorption capability [137]. The results showed that adsorption was unaffected by the presence of other ions, and the composites showed superparamagnetic behavior in the presence of an external magnet.

3.3.2. Cotton cellulose with ferric ion composite

Fe (III) loaded ligand exchange cotton cellulose adsorbent [Fe (III) LECCA] composite was used to create an adsorbent for the selective removal of arsenate from potable water by using column and batch systems. The extraction of arsenate on prepared composite did not depend on pH and notably worked in the pH range of drinking water. Fe (III) LECCA showed high selectivity and a better adsorption rate for removing arsenate from drinking water [138].

3.3.3. Cellulose-carbonated hydroxyapatite nanocomposites

Cellulose-carbonated hydroxyapatite nanocomposites (CCHA) were formed through microwave assistance containing carbonated hydroxyapatite (CHA) nanostructures dispersed in the cellulose matrix using cellulose solution, NaH_2PO_4, and calcium chloride. The maximal adsorption rate obtained was 12.72 mg/g, and the ideal pH range was 4–8 [139]. Adsorption was impeded by PO_4^{3-} and SiO_3^{2-} while the presence of SO_4^{2-} and NO^{3-} did not affect adsorption.

3.3.4. Cellulose-stannic (IV) composite

Cellulose and stannic chloride were mixed in the presence of ethanol with microwave irradiations for 2 m, having 30 s time intervals. In batch experiments, the greatest adsorption capability detected was 16.64 mg/g in a neutral environment. Co-existing ions studies showed that the presence of chloride ions when its presence was ten times the arsenic concentration affected the process of adsorption. The results showed that CSn (Cellulose-stannic) is an outstanding adsorbent [140].

3.3.5. Water bamboo cellulose -FeOOH/CuO composite

As (III) was removed using a composite consisting of water bamboo cellulose. Firstly, cellulose was extracted from water bamboo and then composites were prepared in two steps hydrothermal process. The maximal adsorption capacity observed was 76.1 mg/g in an acidic environment of pH 3.5 [141]. The composite was observed to be eco-friendly, inexpensive, and reusable when it was compared with other adsorbents.

3.3.6. Cellulose-based hydrogels

Hydrogels consisting of glycidyl methacrylate grafted on cellulose having benzoyl peroxide as an initiator were prepared and tested for the elimination of arsenic from water. The hydrogels demonstrated quick and high adsorption efficiency at pH 6 in just 90 min. According to Sips isotherm, the highest adsorption capability was 78.8 mg/g, and almost 100% of adsorption happened at the initial concentration of 25 mg/l [142]. Another study was done in which modified cellulose hydrogels were utilized for the elimination of fluoride and arsenic. The results showed remarkable adsorption efficiency in a very short time (20 min), even at very low concentrations. The maximum adsorption capacity was of arsenate, which was 88.99 mg/g, and the process of adsorption was highly pH dependent [143]. Montmorillonite and cellulose were combined to prepare an effective adsorbent. The results demonstrated that 85% of the total arsenic was adsorbed at pH 6 in 34 min, making the prepared composite a favorable adsorbent for arsenic elimination [144]. Aerogels were prepared using iron oxide nanoparticles and cellulose nanofibrils for arsenic remediation. The maximum adsorption capacity for arsenite was 48 and 91 mg/g for arsenate. The composite was found to be stable in water and showed less mass loss. Moreover, the aerogel showed quality of shape recovery to some extent, which can be useful in transportation purposes [145].

Different cellulose modifications and their adsorption capacities are shown in Table 9. For better adsorption, cellulose is combined with such chemicals that leave toxicity in water, and the efficiency of cellulose-containing adsorbents is highly affected by pH, temperature, reusability, complexity, mechanical strength, and selectivity which should be further investigated to consider cellulose an environmentally sustainable material for arsenic removal [146]. Like sodium alginate, studies have been done on several metals using cellulose ion-imprinted materials; cellulose also needs to be explored in the area of arsenic removal via ion-imprinting technology.

4. Conclusions

The Earth naturally contains significant amounts of arsenic, which often contaminates groundwater.

Table 9. Cellulose composites and their adsorption capabilities.

No.	Kind of cellulose modification	Substance for modification	Adsorption capacity of arsenate	Adsorption capacity of arsenite	pH	Reference
1	Cellulose beads	Iron oxyhydroxide	33.2 mg/g	99.6 mg/g	7	[147]
2	Amino functionalization	Fe(III), amino group, poly(glycidylmethacrylate)-grafted TiO_2	105.47 mg/g	–	6	[148]
3	Phosphorylation and iron loading	Fe(III)	0.94 mol/kg	0.91 mol/kg	7–11 As (III) 2–6 As (V)	[149]
4	Phosphorylation and iron loading	Fe(III)	80% removal	80% removal	7	[150]
5	Iron loading	Fe(III)	1.83 mol/kg	0.24 mol/kg	7	[151]
6	Iron loading	Magnetic iron nanoparticles	2.46 mol/kg	–	2	[152]

This issue has led to elevated arsenic toxicity in water sources across numerous countries. Various methods like electrochemical processes, membrane filtration, adsorption, coagulation, and flocculation have been introduced to counter this contamination. Every method has its limitations, such as complex procedures, requirement of constant care, expensive operation costs and solid sludge production. Among these, adsorption stands out due to its user-friendly nature, cost-effectiveness, and high efficiency. Biopolymers are natural polymers and possess binding sites that can trap heavy metals, like arsenic, within their structures, making them effective chelating agents. This review article highlights the chitosan, alginate, and cellulose-modified adsorbents, exploring their potential for strong adsorption at optimal pH levels. While chitosan-based adsorbents are currently favored in wastewater treatment, broader research into other biopolymers, such as alginate, cellulose, lignin, and keratin, are vital for enhancing arsenic removal techniques. Further research should concentrate on refining biopolymer derivation and modification methods, aiming for cost-effectiveness, biodegradability, reduced toxicity, efficiency, and ease of industrial-scale application. This endeavor paves the way for a sustainable, eco-friendly approach to tackling arsenic contamination.

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