Arsenic Removal Using “Green” Renewable Feedstock-Based Hydrogels: Current and Future Perspectives

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ABSTRACT: In the recent times, scanty access to clean water has been one of the most prevalent problems, affecting humankind throughout the world. This calls for a tremendous amount of research to recognize new methods of purifying water at lower cost, minimizing the use of hazardous chemicals and impact on the environment. The interest of the scientific community in the potential applications of renewable feedstock-based hydrogels for heavy-metal adsorption for water remediation has been continuously increasing during the last few decades. This study is an effort to highlight the application of hydrogels for revolutionizing the present research on heavy-metal adsorption, particularly arsenic. Besides, the arsenic chemistry, health hazards of arsenic to human health, and adsorption of arsenic by natural polymer-based hydrogels have been reviewed in detail. In addition, challenges in taking the hydrogel technology forward and future perspectives like cost, handling, and disposal of the adsorbent have been discussed systematically.

1. BACKGROUND

Water is one of the basic necessities required for the sustenance and continuation of life. However, as a result of rapid urbanization, population growth, and climate disruption, increased demand of clean water has become an unprecedented urgent global issue.1 Some of the common pollutants of water are dyes, phenols insecticides, pesticides, and heavy metals.2 Among the above-mentioned common pollutants, the release of heavy metals into the environment has risen dramatically in the past decades due to rapid industrialization and population growth.3,4

Arsenic ranks first in the top 20 hazardous heavy metals.5 Arsenic is a metalloid and mainly exists in four oxidation states: As(V), As(III), As(0), and As3−; however, the most predominate states are As(III) and As(V).6 The As(III) and As(V) ions are of great interest from research point of view as they are the most common forms of arsenic found in water and cause severe health threats to humankind.7 Contamination of drinking water by arsenic has affected the health of more than 130 million people all over the world, out of which around 110 million people live in Bangladesh, Cambodia, China, India, Laos, Myanmar, Nepal, Pakistan, Taiwan, and Vietnam. According to a report, about 35−77 million Bangladeshi people are at a high risk of exposure to As-contaminated water.8,9

Various strategies have been adopted to remove arsenic from water sources, including oxidation/precipitation, coagulation, ion exchange, and membrane-based separation. These chemical methods are simple, but the disadvantages associated with these methods include huge amount of toxic sludge, slow metal precipitation, poor settling, and aggregation of metal precipitates, which need further treatment before disposal into the environment. Further, fouling of membrane surface, clogging of membrane channels, high operating cost, and more complex operations are several disadvantages associated with the membrane technique.9,10 Hence, there is an imperative need to find novel approaches for purifying water at low cost, using less energy, and above all to minimize the impact on the environment.11 Among all of the aforementioned treatments, the adsorption technique has proven to be more economical and efficient for the removal of even small traces of arsenic from water. Moreover, this method has distinct advantages like easy setup, no need of additional chemicals, and does not produce harmful byproducts. Additionally, this technique requires less operator expertise, low operating cost, and low maintenance cost.12

Of late, hydrogels prepared from biopolymers have been widely used in industrial applications, especially for heavy-metal adsorption. For instance, cellulose, sodium alginate (SA), chitosan, dextrin, and xylan-based hydrogels have been used for the removal of Fe(II), Cu(II), Co(II), Ni(II), Cd(II), As(III), and As(V) from aqueous solutions.13−16 Several biopolymers, such as sodium alginate,17 starch,18 hemicellulose,19 chitin,20 cellulose,21 and their derivatives, have been used to develop hydrogels.

However, several challenges like recovery of nanohydrogel from solution after adsorption and desorption processes, selective removal, recovery of the targeted pollutant, and stability of the hydrogel need to be confronted, which will decide the providence and reusability of the bio-based hydrogel. In addition, the reusability of the hydrogel by desorption of
heavy metals and regaining of the adsorption capacity after several treatment cycles need to be considered. Such findings are encouraging because if a hydrogel prepared from renewable feedstock is highly reusable, it can make the overall process more economical and sustainable.22

Several review articles have been published to date, which deal with arsenic removal by adsorption, but the present study endeavors to highlight the application of hydrogel as an adsorbent (mainly renewable resource-based hydrogels) for arsenic removal. Herein, we attempted to summarize the scientific roadmap of various strategies used for arsenic removal, with special focus on renewable feedstock-based hydrogels. The discussion covers arsenic chemistry, resources of arsenic, hazards of arsenic to human health, various adsorption technologies for arsenic removal, general discussion of hydrogels, and adsorption of arsenic by hydrogels in detail. Besides, the aim of this study is to compile and represent the adsorption potential of various hydrogels for arsenic removal prepared from pectin, chitosan, cellulose, sodium alginate, and cellulose. A summary of published research data with some results has also been presented. Further, challenges like operational pH range, interference, and hydrogel recovery are discussed. Besides, reusability of hydrogels, stability and resource recovery, safe disposal of arsenic sludge, and future perspectives of hydrogel are discussed in this study.

2. ARSENIC REMEDIATION BY HYDROGEL

Recently, hydrogel-based adsorbents have attracted special attention because of their potential in removing heavy metals from wastewater.19 Hydrogels are three-dimensional porous structures that exhibit chemically responsive functional groups, enabling the hydrogel to readily capture metal ions from contaminated water and simultaneously release and clear these metal ions from the hydrogels upon changing the environmental conditions. Owing to the presence of carboxylic acid, amine, hydroxyl, or sulfonic acid groups, metal ions can penetrate the porous network structure of hydrogels and form stable complexes with these groups.23 Arsenic remediation by hydrogels involves several mechanisms like electrostatic interactions, surface area and pore volume formation of hydrogel, complex formation, chelation, and physiosorption.24−27

However, it is very difficult to comment on the type of mechanism involved in arsenic removal by hydrogels. pH is also an important parameter that affects hydrogel performance by influencing metal-ion chelation with the functional groups of hydrogel.28 Perhaps, the most predominant factor is the simultaneous presence of numerous metal ions and counterions, which may participate in complex synergistic and conflicting mechanisms as they compete for binding sites in the polymeric hydrogel structure. It is evident from the literature that different mechanisms are involved in arsenic removal by different hydrogels. According to the literature, because of the presence of a large number of polar functional groups, electrostatic interaction is considered to be the main mechanism for the removal of arsenic ions.29,30 Moreover, the hydrogel having negatively charged surface can adsorb positively charged metal ion through electrostatic interaction mechanism. Besides, in some cases, chelation of heavy metal with the polar groups of hydrogel has also been reported. The ion-exchange mechanism of arsenic removal has also been reported by the hydrogels possessing acidic or basic functional groups.22

2.1. Chitosan. Chitosan is the second most abundant polymer in nature after cellulose. Chitosan is poly[β-(1→4)-2-amino-2-deoxy-D-glucopyranose], which is formed by β(1−4)-linkages between D-glucosamine and N-acetyl-D-glucosamine units. Chitosan exhibits high affinity for heavy metals like mercury, arsenic, cadmium, lead, etc. Adsorption of heavy metals can occur by chelation, electrostatic interaction in neutral solutions, or ion-exchange mechanisms in acidic solutions.31,32 At neutral pH, when the −NH₂ groups are not protonated, chitosan can undergo the following reaction with the metal ions (1)

$$M^{n+} + RNH_2 \leftrightarrow M(RNH)_2^{n+} \quad (1)$$

From the above equation, we observe that the efficiency of chitosan as an adsorbent increases with the increase in the number of free −NH₂ groups and hence also the degree of deacetylation.33

Chitosan and its derivatives have been widely used for arsenic removal since long time.34 For instance, Chen et al. synthesized chitosan beads using agricultural waste for arsenic removal with maximum adsorption efficiency at pH 5. The synthesized chitosan beads favored the adsorption of As(V) (1.83 mg/g) over As(III) (1.94 mg/g); however, the authors did not explain the reason for the same behavior. Su et al. fabricated highly porous nanoscale zero-valent iron/chitosan composite foams (ICCFs) for inorganic arsenic removal by freeze-drying method (Figure 1). The fabricated ICCFs exhibited good mechanical properties as well as excellent removal efficiencies of As(III) (114.9 mg/g) and As(V) (86.87 mg/g). The authors discussed the mechanism of arsenic removal after As(III) removal.
removal in detail and revealed that the adsorption follows a pseudo-second-order model and their isotherms follow the Langmuir adsorption model.

Molybdate-impregnated chitosan beads for effective adsorption of As(V) were prepared by Dambies et al. They observed that maximum adsorption of As(V) occurred in the pH range of 2–4 with a minimum release of molybdate ions. Boddu et al. used composite chitosan biosorbents (CCBs) for arsenic removal. Besides, the effect of pH on the adsorption capacities of As(III) and As(V) by CCB was investigated in detail.

Iron chitosan composite hydrogel was used for the removal of As(III) and As(V) at pH 7.0 under equilibrium and dynamic conditions:

\[
\text{Adsorption per gram of adsorbent (mg/g)} = \frac{q_e}{m} \times \left( \frac{C_0 - C_e}{C_0} \right)
\]

where:
- \(q_e\) is the adsorption capacity at equilibrium (mg/g).
- \(C_0\) is the initial concentration of the metal ions (mg/L).
- \(C_e\) is the equilibrium concentration of the metal ions (mg/L).
- \(m\) is the mass of the adsorbent (g).

**Figure 2.** (a) Equilibrium adsorption isotherms and (b) linearized Langmuir plots for As(III) and As(V) by ICB and ICF. Conditions: concentration of As(III)/As(V), 1–10 mg/L; pH, 7; dose rate, 5 g/L; total aqueous volume, 20 mL; stirring time, 4 h. Reprinted from ref 36 Copyright (2009), with permission from Elsevier.

**Figure 3.** (a) Schematic diagram of MBOP column study for the removal of As(III) from water. (b) Vibrating sample magnetometer magnetization curve of MBOP before and after adsorption. Reprinted from ref 38 Copyright (2017), with permission from Elsevier.

**Figure 4.** Scanning electron microscopy images of MBOP (a) before and (b) after As(III) adsorption. Reprinted from ref 39 Copyright (2011), with permission from Elsevier.
conditions. The effect of other anions on the adsorption of arsenic was also evaluated, and it was shown that the presence of sulfate, phosphate, and silicate ions did not cause interference in the adsorption behavior of arsenate/arsenite. The equilibrium data were fitted to the Langmuir adsorption model, and the monolayer adsorption capacities of iron chitosan flakes (ICFs) calculated from the Langmuir model were 22.47 ± 0.56 and 16.15 ± 0.32 mg/g for As(V) and As(III), respectively, which are considerably greater than those of iron chitosan granules (ICBs) 2.24 ± 0.04 mg/g for As(V) and 2.32 ± 0.05 mg/g for As(III) (Figure 2).36

Chitosan has also been used for the preparation of superparamagnetic magnetic binary oxide particles (MBOPs) via template method for As(III) removal. The column study was done at two different flow rates (2.0 and 5.0 mL/min). A schematic diagram of the column study for the removal of As(III) from water is shown in Figure 3a. It was observed that the adsorption rate constant (K) and adsorption capacity coefficient (N) were calculated through the logit method37 to be 0.2076 (L/(mg h)) and 1015 (mg/L), respectively, for the 5.0 mL/min flow rate. Under both dry and wet conditions, MBOP get attracted toward the magnet, which confirms their magnetic nature, as shown in Figure 3b.38

The same group also synthesized magnetic binary oxide particles (MBOPs) using chitosan template for uptake capacity of As(III). It was demonstrated that the maximum uptake of As(III) was at pH 7.0 and that the equilibrium data were well fitted to the Langmuir and Freundlich isotherms. No change in the surface morphology was observed before and after arsenic adsorption (Figure 4), revealing the physical adsorption of arsenic on the adsorbent.39 Miller et al.40 prepared TiO2-impregnated chitosan beads (TICBs) for arsenic removal. They revealed that the adsorption capacity of TICB was influenced by solution pH, TiO2 loading, and exposure to UV light.

2.2. Sodium Alginate. Sodium alginites (SA) are naturally derived polysaccharides obtained from brown macroalgae composed of (1-4)-linked β-D-mannuronic acid and α-L-guluronic acid monomers. Divalent metal ions are used to cross-link SA for the fabrication of SA hydrogels. SA-based sorbents are found to be very effective in removing toxic heavy
metals, such as Ni, Cu, Zn, Cd, Hg, Pb, Nd, Eu, Yb, and Ra, from water. Besides, SA microbeads have a great potential for arsenic removal and can be beneficial for developing countries.

Akaganeite nanorods of uniform size were prepared by Cho et al. by hydrolysis of ferric ions in a two-phase system and encapsulating the same in alginate microbeads for arsenic removal (Figure 5). It was found that the arsenic removal efficiency could be affected by the akaganeite amount, treatment time, pH, and microbead size. The lower adsorption of As(III) relative to As(V) by alginate microbeads can be corroborated to the uncharged nature of As(III), which led to weaker interactions with positively charged akaganeite. Nonetheless, the mechanism of arsenic removal and reusability of the synthesized hydrogel was not discussed.

El-Sherbiny et al. developed calcareous soil–alginate composites for adsorption of Fe³⁺, Mn²⁺, and As(V) from water. The composites showed the highest removal efficiency for Fe³⁺ (100%), followed by Mn²⁺ (89%) and As(V) (50%). However, the authors did not discuss the effect of various parameters like pH, temperature desorption, and reusability of the prepared composite hydrogel. Vu et al. reported the synthesis of alginate beads for As(V) removal by encapsulating magnetic graphene oxide (mGO), as shown in Figure 6. Besides, the authors explored the role of cross-linking metal cations in pH control. The study revealed that As(V) adsorption was not significantly affected by acidic or basic pH values because of the presence of released Ca²⁺, which controlled the local pH when target solution penetrated the beads.

Ocinski et al. examined the adsorption of chemically pretreated water treatment residuals (WTRs) with calcium alginate polymer for As(III) and As(V). They studied the effect of pH on arsenic adsorption by WTR-loaded alginate, and the results revealed that the adsorption efficiency decreased for both arsenite and arsenates at pH 7. The adsorption mechanisms of As(III) and As(V) on Fe–Mn binary oxides are presented in Figure 7. Sarkar et al. synthesized low-cost ferric hydroxide microcapsule-loaded alginate beads as an adsorbent for arsenic removal.

Taken together, the above sections clearly indicate that the effective adsorption of arsenic can be achieved by using chitosan- and sodium alginate-based hydrogels. However, the mechanistic insights involving magnetic hydrogels are still in the early stage of development. Further, much effort should be made in this field to prepare highly regenerated hydrogels selective toward a particular metal ion in a mixture of other ions with well-defined mechanism action.

2.3. Other hydrogels Used for Arsenic-Contaminated Water Treatment. Natural polymers like pectin, cellulose, and hemicellulose are also being used by several researchers for arsenic removal. For instance, Fe(II)-loaded pectin hydrogels were prepared for the adsorption of As(V) from water using three different cross-linkers. The hydrogels cross-linked with 5% glutaraldehyde showed the maximum uptake of both Fe(II) and As(V). The effect of pH and temperature on the adsorption of As(V) was also evaluated. The maximum adsorption was observed at 35 °C and pH 7.0—9.2.

Cationic-modified cellulose hydrogel for fluoride and arsenic adsorption was reported by Meng et al., and the results revealed that the modified hydrogel exhibits higher adsorption efficiencies for F⁻, AsO₃³⁻, and AsO₄³⁻ simultaneously from aqueous solution. Anirudhan et al. synthesized iron(III)-coordinated amino-functionalized poly(glycidyl methacrylate)-grafted cellulose for arsenic(V) adsorption. Hemicellulose-based hydrogels and their application for the removal of arsenic and chromium ions were also described in the literature.

 α-Fe₂O₃-impregnated chitosan beads using As(III) as imprinted ions were prepared for the adsorption and removal of As(III) ions from aqueous solutions. In another research, Fe and Mn oxides (FMBO) immobilized into a konjac glucomannan (a kind of polysaccharide)-based aerogel matrix were prepared to remove As(III) from water.

3. CHALLENGES OF THE CURRENT STRATEGY

Despite the promising future of the bio-based hydrogels in water remediation, several challenges need to be addressed at the bench scale so that the strategy can be transferred from lab to commercial practice. For instance, the effect of pH on the adsorption efficiency of hydrogel is an important parameter, as this leads to changes in the surface properties of the hydrogel and the degree of ionization of the adsorbate molecule. The pH of zero point charge (pHZPC) corresponds to the pH value at which the surface charge density of an adsorbent molecule is zero. pHZPC plays a significant role in explaining the efficiency of the adsorption process in terms of ion attractions from oppositely charged surfaces. At pH < pHZPC, the adsorbent exhibits positively charged surface, and at pH > pHZPC, the surface of the adsorbent exhibits negative charge. The effect of pH on the adsorption efficiency of heavy metal by hydrogels has been scrutinized by some of the research groups. Mostly, hydrogels perform in a very small pH range and hence that particular pH has to be adjusted so that maximum pollutants can be remediated. However, this study can raise a question on the stability and performance of bio-based hydrogels in consecutive cycles. Therefore, to reuse bio-based hydrogels, consideration of the operational pH range is an important parameter.

Besides, high adsorption rate is a significant factor that needs to be considered from a practical point of view. The bulk hydrogel takes longer equilibrium time compared to smaller hydrogel particles. In one report, comparison of bulk hydrogels and small hydrogels was studied, which revealed that smaller hydrogel particle size decreased the adsorption time from 2 h to 5 min as the latter have high surface area, with more exposed
site for adsorption of heavy metal.\textsuperscript{56} However, if the hydrogel particle is too small, its recovery from the solution is another challenge. The separation of tiny hydrogel particles through conventional techniques, such as sedimentation, is ineffective, whereas other methods, such as centrifugation, are energy-consuming. Recently, it has been demonstrated that if the hydrogels are embedded with magnetic nanoparticles, they can be separated from the solution magnetically.\textsuperscript{22}

The hydrogel performance in the presence of different metal ions is another hindrance for the commercialization of bio-based hydrogel. The adsorption efficiency of the hydrogel decreases in the presence of other ions because of the competition created by other metal ions. Additionally, if other ions like calcium, chlorides, natural organic matter, sodium, and sulfates are present in higher concentration, they will compete and adsorb more onto the hydrogel surface and hence make the recovery process of economic metal ion very tedious. Hence, a decrease in the adsorption amount of targeted metal ions increases the regeneration requirements and poses difficulties in situations where only selective removal of single metal ion is desired. The selectivity of a hydrogel toward particular metal ions can be made by introducing specific functional groups. The selectivity of the targeted metal ion can be enhanced by the introduction of functional groups capable of forming hydrogen bonds with the targeted metal other than the ions present.\textsuperscript{22} However, this area should be explored more regarding the regeneration and reusability of hydrogels than hydrogel functionalization.

In addition, recovery of heavy-metal ions from hydrogel structure, especially those having high market value, such as nickel, copper, platinum, gold, phosphorous, etc., should be taken into account. After recovery, the aforementioned metals can be reused, for example, recovered chromium metal ion can be used in steel manufacturing, metal finishing, chemical industries, and so forth. Similarly, recovered phosphorous ion can be utilized in the fabrication of fertilizers, detergents, and corrosion inhibitors. For this, magnetic iron oxides method is desirable because of its cost effectiveness and less energy requirement, but this method is found to be effective only in acidic environments.\textsuperscript{57,58} Nonetheless, recovery of arsenic metal ions from hydrogel is not profitable because of the hazardous effect on the environment. Hence, recovered arsenic metal ions should be encapsulated through solidification/stabilization processes with long-term stability and minimal leaching effect, which makes the toxic waste safer prior to arsenic disposal.\textsuperscript{59}

Finally, the research should be concentrated on reusing the bio-based hydrogels by desorbing the heavy metals and retrieving the adsorption capacity in consecutive cycles so that the complete treatment becomes extra cost-effective and sustainable. Mainly, adsorption of pollutants is achieved by two types of reactor: (i) fixed-bed reactor (hydrogel in particle form) and (ii) stirred tank reactor (hydrogels suspended in solution). However, fixed-bed reactor is not a good choice because of clogging, compaction, and pressure drop problems, whereas in stirred tank reactor, separation of the hydrogel from the solution after adsorption or desorption process is a major challenge.\textsuperscript{60} Again the separation of hydrogel depends on the particle size of the same. If the hydrogel particles are of micro or nanoscale, then the separation is difficult through conventional techniques, and the centrifugation process makes it energy-consuming and costly. For the recovery of hydrogel particles from the solution, fabricating magnetic hydrogels is more feasible because it is highly selective, fast, and environmentally friendly compared to other techniques. Various kinds of iron oxide nanoparticles like Fe$_3$O$_4$ and γ-Fe$_2$O$_3$ exhibit high saturation magnetization values and hence can be used for the synthesis of magnetic hydrogels.\textsuperscript{22} However, nanosized magnetic hydrogel particles need stronger magnetic field compared to microsized particles because of several influences taking place in nanoform like drag, gravitational, buoyancy, etc. Moreover, as particle size increases, the adsorption efficiency decreases because of the decrease in the surface-to-volume ratio. Hence, the optimum magnetic hydrogel particles should be fabricated to maintain the balance between the adsorption capacity and magnetic separation.\textsuperscript{51,52} A schematic representation of the overall challenges that need to be focused for the commercialization of the biopolymer hydrogel is given in Figure 8.

4. CONCLUSIONS AND FUTURE PERSPECTIVES

Nowadays, energy and water are the two most important challenges that we face in our society. Sustainable, low-cost supply of clean water to the society is crucial for global prosperity and health. Adsorption technology is expected to
play an important role in water remediation because it is both cost-effective and energy-efficient. To achieve this, biopolymer-based hydrogels is the most promising alternative to the conventional sorbents for removing one of the main contaminants, arsenic, from water. A new avenue can be opened for solving the environmental and energy problems by using the natural polymer hydrogels for heavy-metal adsorption to maintain social sustainable development. Hydrogels prepared from SA, pectin, chitosan, and cellulose have attracted attention from both academic and industrial points of view as these polymers are less expensive, bio-compatible, and biodegradable in nature. Besides the comprehensive “green” utilization of these materials, it also provides the possibilities of preparation of various advanced materials, which can replace the synthetic polymers. The future of hydrogels in water remediation is somewhat bright but needs the collaborative efforts of both government and nongovernment sectors to develop some sustainable and scale-up innovative approaches to supply clean drinking water to the society. This perspective article connotes that this field of research has enough space, which should be covered in the near future, and hopefully, the laboratory-scale research should be transferred to industrial scale. Further, smart hydrogel materials can act as a new generation of adsorbing material by incorporating functional nanomaterials, for example, graphene oxide (GO), carbon nanotubes, and so forth. While fabricating hydrogel-based adsorbent, several important parameters need to be addressed, such as pore size, surface charge, and understanding the interactions between adsorbent and adsorbate. Scientists should develop hydrogels with high selectivity toward a particular metal ion besides recovering the adsorbed metal for reuse in various sectors. In addition, a hydrogel should be stable in consecutive cycles of treatment from economic and sustainability points of view.

To this end, disposal of arsenic waste is a major problem in developing countries like India. It is also one of the main barriers for the successful implementation of available technologies for arsenic removal. Ideally, arsenic should be disposed in an environment without any further risk of environment contamination. Available methods for arsenic disposal are landfills, cow dung mixing, stabilization, anaerobic digesters, and soil disposal. Future research should be directed to develop some innovative methods that may provide a final solution for the arsenic waste disposal problem even in nonlandfill areas. In this context, green hydrogels have promising future and should be researched upon in this direction to cater safe and clean drinking water to our ever-growing population.

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