

Approaches of Arsenic Removal from Water Bodies: Application and Limitations

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Abstract The literature outlines numerous traditional methods used to eliminate arsenic. New technologies have been employed to enhance the removal process, which is more realistic, cost-effective, and eco-friendly. This study reviews the latest technologies, along with the fundamental principles they are based on. Traditional arsenic removal methods were compared with their newer counterparts to assess their effectiveness and suitability for various groundwater sources. The discussion centers around unique methodologies, the water source sampled, the technology utilized, and the impact of other factors. Further, these approaches were evaluated based on their environmental impact and economic feasibility. The use of some innovative technologies is still restricted to laboratory conditions, which makes it difficult to calculate their removal efficiency under natural conditions accurately. Traditional approaches necessitate several processes to achieve the desired concentration. Additionally, only filtration can reduce the arsenic content to a specific level in an area with existing iron. Furthermore, electrocoagulation technologies can be utilized only in locations where electricity is readily accessible and cost-effective. The innovative technologies in this study showed greater removal efficiency compared to the traditional methods. Although the expenses of these technologies are not applicable in all circumstances, they are recognized as environmentally friendly concepts. These technologies effectively eliminated arsenic without the need to alter the pH or oxidation state of the contaminated water.

Keywords Arsenic Contamination, Arsenic Removal, Water Quality, Water Treatment Methods

1. Introduction

Arsenic (As), a highly toxic element, occurs naturally in groundwater due to the solubility of certain compounds [1], [2]. Widely termed "ubiquitous," arsenic permeates various environmental compartments, including minerals, rocks, soil, water, plants, and animals [3]. Ranking twentieth in abundance within Earth's crust [4], arsenic manifests as a colorless, odorless, and tasteless substance, detectable only through chemical analysis. Anthropogenic activities such as mining, combustion engines, and the use of arsenic pesticides contribute to its presence alongside naturally occurring sources [1, 5].

Both natural processes and human activities contaminate groundwater with arsenic [6, 7]. Arsenic primarily exists in inorganic forms in water, with arsenate (V) (H_2AsO_4^-) and arsenite (III) (H_2AsO_3^-) being the predominant anionic species. Arsenite is more toxic than arsenate, with arsenic (III) compounds exhibiting 60 times greater toxicity than arsenic (V). Furthermore, groundwater typically contains 67-99% arsenic (III) compounds, necessitating the removal of both forms [8].

The speciation of arsenic in water is influenced by factors such as pH, redox potential, ion presence, and microbial activity. Their removal from water poses significant challenges due to the unstable nature of arsenic compounds [4].

Arsenic poses grave health risks and is recognized by the USEPA as the sole carcinogen transmitted through drinking water, contributing to various cancers, including

lung, bladder, skin, liver, and kidney [9]. Over 150 million individuals across 70 nations face arsenic-related health concerns, with concentrations exceeding 100 to over 2000 µg/L in affected regions [5,7]. Regulatory bodies, including the Environmental Protection Agency (EPA) and the World Health Organization (WHO), have set a maximum allowable limit (MAL) of 10 µg/L for arsenic in drinking water [8], thus underscoring the need for efficient removal techniques. Similarly, Canada and Ontario Province also regulate this concentration [10].

Traditional approaches to removing arsenic from drinking water encompass several methods, such as adsorption/filtration, coagulation/filtration, activated

alumina lime, softening, reverse osmosis, and ion exchange, as outlined by Health Canada [10]. Among these, the predominant method involves the utilization of various sorbents, as indicated by Amen et al. [11] and Ardaul et al. [12]. Additionally, employing chemical agents such as ferric chloride in the coagulation process, followed by flocculation and filtration, stands out as a widely utilized technique for arsenic removal, as Mohindru et al. [13] highlighted. Table 1 provides a comparative analysis of the efficiency of these conventional techniques in arsenic removal. A schematic diagram illustrating the procedure of arsenic removal treatment is shown in Figure 1.

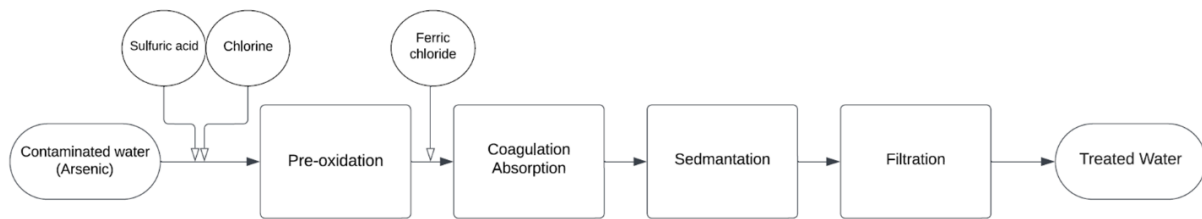


Figure 1. Flowchart of arsenic removal processes. [10, 11]

Table 1. Arsenic removal techniques using conventional technologies

Technology	Chemical reagent	As (V) Removal efficiency (%)	As (III) Removal efficiency (%)	Ideal conditions	Reference
Ion exchange	Anionic resins	80-95	<30	-	[20]
Adsorption	Activated carbon or activated alumina	>95	30-60	pH 5.5-6	[7,21,27,28]
Coagulation	Ferric chloride	90-95	<30	pH 6-8	[15-18]
	Aluminum, copper				

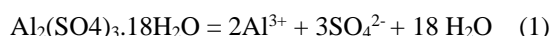
2. Conventional Arsenic Removal Techniques

2.1. Coagulation

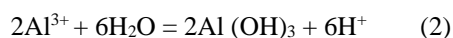
In the coagulation process, ferric or aluminum ions are added to a solution from which desired ions must be removed, which, in our case, is arsenic ions (arsenate or arsenite). The fine particles of arsenic then aggregate into coagulates as the absolute values of the zeta potential of arsenic particles are strongly reduced by ferric or aluminum ions. Finally, arsenic ions precipitate and coagulate with ferric or aluminum ions, which are removed from water through filtration [14].

The chemical reaction of the coagulation process (alum with As (V)) is demonstrated below:

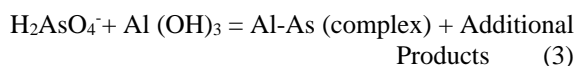
Step 1: Alum dissolution



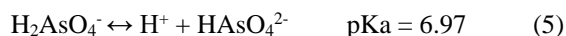
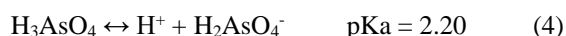
Step 2: Aluminum precipitation (acidic)



Step 3: Co-precipitation (Non-stoichiometric)



Coagulation is an effective process for arsenic removal, followed by other processes such as filtration and microfiltration [15]. Its effectiveness highly depends on the pH and the amount of coagulant used [16]. A similar reaction occurs when ferrous ions (Ferric Sulphate or Ferric Chloride) are added as coagulants. They also produce Fe-As complex and other end products. The acid-base detachment reactions of arsenic acid are displayed below:



Ferric and aluminum ions are traditionally used as coagulants [17]. Ferric compounds have been used in most of the research because they are more effective in removing arsenic from water than others. Ferric ions have shown a noticeable range of arsenic removal without the addition of any coagulant [15]. However, for ferric ions to function properly, the pH of the water needs to be lower, which requires the addition of acid [16]. For example, if the pH of tested water is 7, HAsO_4^{2-} would be the dominant species present [15].

Coagulation is popular in developing countries due to its low cost and effectiveness in reducing arsenic content to as low as 10.0 $\mu\text{g/l}$ [18]. However, it cannot be performed in isolation and requires other processes such as filtration to remove particles and achieve further arsenic removal. Another limitation is that oxidation is needed to convert from As (III) to As (V). Further, the membrane has fouling

problems [16].

Electrocoagulation is a relatively new technology that uses electrodes to produce in-situ coagulants. Both fields and experiments show a 99.9% removal efficiency of this process [8].

2.2. Ion Exchange

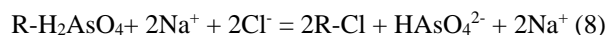
This removes the contaminant or desired ion from the contaminated water by running it through an ionized solid phase. The solid phase is made of a synthetic resin chosen according to the contaminant ion type that needs to be removed. Up or down feed water flow is maintained until the resin is exhausted or filled with contaminated ions. Once the resin is exhausted, it must be regenerated to enhance removal efficiency [14].

The ion exchange and regeneration reactions are demonstrated below [19].

Ion Exchange:



Regeneration:



Here, R is ion exchange resin. This process is also affected by a defined pH (6.5-9) range, outside which the arsenic removal efficiency decreases [14]. The pH effect of As (V) and As (III) can be described by the following graph [20]. In Figure 2, As (V) species H_2AsO_4^- and HAsO_4^{2-} are the predominant species with pH ranging between 2.2 and 11.6. AsO_3 becomes predominant after pH becomes 11. In Figure 3, Arsenic (III) species H_3AsO_3 is predominant when pH is less than 9.2. Other Arsenic (III) species are dominant at a higher pH ($\text{pH} > 10$) [20].

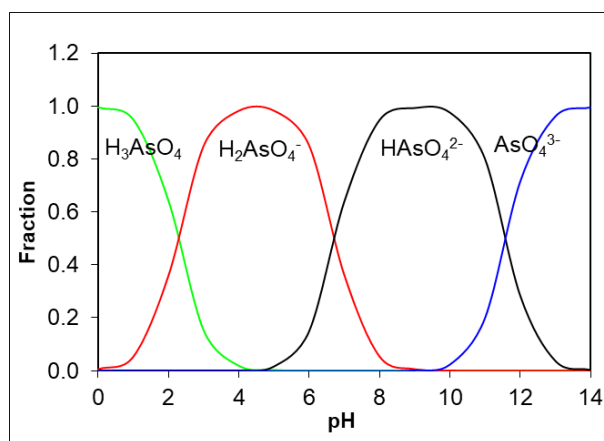


Figure 2. Fraction of As (V) species vs pH. [20]

Ion exchange for arsenic removal is applicable only when the water has a low TDS (Total Dissolved Solids) and low sulfate concentration. The presence of Fe (III) ions can form complexes with arsenic, which cannot be removed by ion exchange. Only strong base resins are used for arsenic removal in the ion exchange process. The main problem

with chloride resin is that it increases chloride concentration in treated water, resulting in corrosive water. Resins also face fouling problems caused by the scaling of minerals such as Ca and Fe.

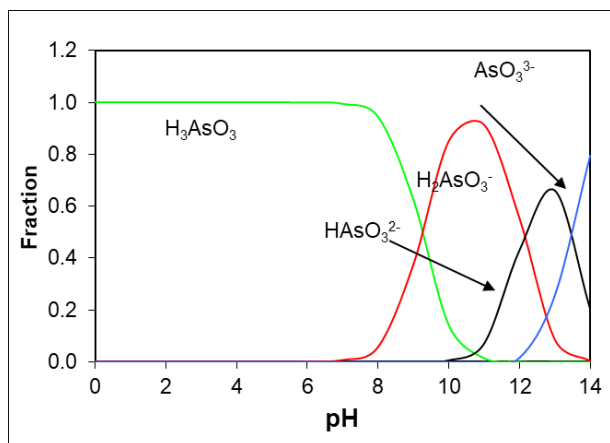


Figure 3. Fraction of As (III) species vs pH. [20]

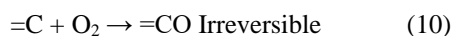
2.3. Adsorption Technologies

In adsorption, a solid or liquid surface is called an adsorbent. A gas or liquid solute, often the solution with the contaminated ions, accumulates on the surface of the adsorbent, creating a molecular film called adsorbate.

Physical Adsorption:



Chemical Adsorption:



Surface Adsorption:



Adsorption is faster and more effective than coagulation. Almost for all treatment technologies, the oxidation state of arsenic plays an important role because As (V) is much more easily adsorbed than As (III) [14]. As a result of the

toxicity and difficult removal process of arsenic (III), it must be oxidized before or during the adsorption process [21]. Oxidation is usually done by iron (0, II, VI), manganese (IV), manganese (VII), chlorine, or hypochlorite. Numerous adsorbents for arsenic removal from water have been established, including materials like activated alumina and iron hydroxide-coated alumina [7].

Ferrihydrite is a common adsorption agent. Studies have shown that in-situ use of ferrihydrite removes more arsenic than prepared ferrihydrite. Granular ferric hydroxide (GFH) is another arsenic removal agent [21]. The process using this agent is temperature-dependent, as studies indicate that the adsorption of arsenic by GFH is a natural endothermic procedure. Goethite, a natural iron oxide, is an adsorption medium for arsenic removal. A study shows that hematite (another iron oxide) and goethite could strip off 80% of arsenic from water under any pH range, regardless of the initial concentration of arsenic. Aluminum hydroxides are more effective in removing As (V) than As (III) [21]. Layered double hydroxides (LDH) are also well-known arsenic removal agents, for which the "memory effect" mechanism is vital. This mechanism can regain structure during rehydration. The order of ions disturbing arsenic adsorption is phosphate > humic acid (HA) > silicate > sulfate [21].

3. Innovative Arsenic Removal Techniques

Numerous articles and research have investigated different removal approaches for arsenic, and they all focus on reducing the footprint and the economic side of the treatment, taking in their prospective high removal efficiency [22, 23, 25-27, 34-40]. Table 2 summarizes the removal percentage of the techniques that have been discussed.

Table 2. Comparison of the arsenic removal efficiency in novel techniques

Treatment name	Dosage	Dosage concentration	Initial arsenic concentration	Removal efficiency	Reference
Arsenic sorption properties of Mg-Al sulfate layered double hydroxides (LDH)	Mg-Al (LDH)	100 mg/L	100 µg/L	99.99%	[25-27]
Cupric oxide	CuO	0.5 g	12.4-398 µg/L	86-100%	[28]
Pretreated waste tea fungal biomass	FeCl ₃ pretreated and autoclaved fugally.	1.0 g/50 mL	1.3-0.9 mg/L	100% of As (III) 77 % of As (V)	[35]
Nano scale zero-valent	NZVI	1.0 – 1.5 g/L	10 -100 mg/L	99%	[34,36,38,40]

3.1. Arsenic Removal via Layered Double Hydroxide Adsorbents

This technique has been investigated since 2005 [24-27]. Chetia et al. [26] established a hydroxalcalite-like compound known as layered double hydroxide (LDH) to be an efficient adsorbent for arsenic exclusion. It has a high anion exchange capability and high surface area. This study conducted a batch experiment using adsorption based on a calcination-rehydration reaction. It further stated that factors like pH, dose of adsorbents, and shaking time impact removal efficiency.

LDH is anionic clay and is found as a natural mineral. The relatively weak interlayer bonding structure of LDH allows the capturing of organic and inorganic anions. Zn-Al LDH can regain structure during rehydration, also known as the "memory effect." Moreover, two environmentally friendly non-heavy metals, Zn and Al, are presented in a secured hydroxide framework. LDH can be regenerated with thermal treatment in hydroxide and anionic solution. This process is highly pH-dependent, as it affects the adsorbent's surface charge and the degree of ionization speciation of the adsorbate.

Removal efficiency of 99.99% was achieved from a 100 µg/L of arsenic solution. A 100 mg/L of Zn-Al LDH solution was used to get this removal efficiency. It was subjected to 1.5 hours and 200 RMP. This study was done in a laboratory using a synthetic arsenic solution, so no effects of competitive anions or other elements are mentioned.

3.2. Arsenic Elimination from Groundwater Utilizing Cupric Oxide

The utilization of cupric oxide has been investigated for the removal of arsenic [28]. A study was conducted on groundwater samples from local countryside wells in the western United States [29]. Approximately 46 groundwater samples were collected to observe the capability of cupric oxide (CuO) elements to affect arsenic elimination in a natural environment. This research did not include any adjustment on pH or oxidation, and no competitive anions were added or removed. Only a one-step procedure removed the arsenic from the collected samples.

Conventional arsenic adsorbents have limitations regarding pH adjustments, oxidation conversion, preference of oxidation state, and competition with other anions. It was established that cupric oxide is an efficient adsorbent that overcomes all the mentioned limitations [30]. The high adsorbent capacity is achieved because of its higher surface area, allowing more adsorption. Like other adsorbents, the cupric oxide can be regenerated using a sodium hydroxide solution and reused to remove arsenic.

Two US patents have been received for this discovery [29]. The study was done on groundwater, and 46 samples were tested to observe the performance of cupric oxide on arsenic removal. Treatment with cupric oxide showed very

little effect on the pH. Only in two samples where the pH was much higher (close to 9) could cupric oxide lower them slightly (around 8.5). Other samples' pH remains within the valid limit (6.5-8.5) before and after treatment. Unlike other adsorbents, competitive anions have no significant effect on arsenic removal. Moreover, the concentration of main elements, for instance, Mg, Ca, Na, Cl⁻, K, SO₄²⁻ and some trace elements, such as Cu, Fe, Mn, Cr, and Pb, stayed nearly unchanged due to the treatment process. Only concentrations of silica and phosphates were affected but remained within acceptable limits.

The initial arsenic concentration in the 46 well water samples ranged from 12.4 to 398 µg/L, and the final concentration ranged from 0.1 to 29.1 µg/L, which indicates a removal efficiency of 86-100%.

Phosphate has shown a significant impact on arsenic removal. A high concentration of competing anions was tested, and results showed that even with a high concentration of phosphates, cupric oxide could eliminate 83% of As (III) and 100% of As (V). CuO has high ZPC, and it is competent for oxidizing arsenic [As (III) to As (V)]. These are the two main reasons behind this high removal efficiency.

3.3. Biological Methods of Arsenic Removal from Water

Due to the complex nature of arsenic compounds, applying a solo biological method is challenging. Nonetheless, bacteria can oxidize and reduce arsenic compounds in aquatic environments [31,32,33]. Recently, research by Islam et al. [34] coupled a green microalga with nano-zero valent iron as an adsorbent. In another study, tea fungus (byproduct from black tea fermentation) has been experienced for its isolation capability of arsenic As (III), As (V), and Fe (II) from groundwater samples collected in West Bengal, India [35]. Two different materials were applied: an autoclaved tea fungal mat and a ferric chloride pretreated fungal mat. The research was conducted in Kolkata, West Bengal, India. For this report, only arsenic species will be recognized.

The findings showed that the biosorption rate is inclined to be directly proportional to contact time and adsorbent dosage. Moreover, the samples' average concentrations of As (III) and As (V) were 1.30 mg/L and 0.90 mg/L, respectively. The removal efficiency for both the pretreated (FeCl₃) and autoclaved fungal mats was 100% of As (III) after 0.5 hr contact time and 77 % of As (V) after 1.5 hr contact time, and the optimal dose was 1.0 g/50 mL. This dosage was attained after batch mode experiments via agitating different concentrations of Adsorbent with the following concentrations (0, 0.5, 1.0, 1.50, 2.0, and 2.50 g/50 mL).

Many arsenic removal techniques are based on chemical approaches, which utilize various chemical reagents; however, the researchers wanted to investigate an eco-friendly approach using a biological waste byproduct.

Nevertheless, they did not neglect the economic side, which they claimed is less expensive than many other removal methods that have been used. Moreover, conducting this experiment in a well-recognized area of severe arsenic contamination gives this research particular credibility because it monitors actual arsenic-contaminated groundwater.

3.4. Arsenic (V) Removal Using Nanotechnology

Several studies were conducted on the elimination of arsenic (V) from groundwater using nanoscale zerovalent iron [36-40]. Amongst the most broadly employed nanoparticles, nano-zerovalent iron (nZVI), maghemite (γ -Fe₂O₃), and magnetite (Fe₃O₄) nanoparticles have been researched for the treatment of polluted water [2,18]. The iron-nanoparticles-based coupled with ion exchange techniques were reported to be an expansive method [39]. However, it can be more cost-efficient since it can be reused after regeneration. Still, the drawback is that it is a sludge-producing process requiring further downstream treatment.

In West Bengal (India), a synthetic nanoscale zerovalent (NZVI) was investigated to explore the effect of adsorption kinetics, pH, sorption mechanism, and anionic impact on arsenic removal efficiency [40]. This research utilized advanced analyzing apparatuses: field emission scanning electron microscopy (FE-SEM) and high-resolution transmission electron microscopy (HR TEM). Furthermore, the research recognized the sorption mechanisms through conjoining different microscopic and spectroscopic research in the same field. Also, it investigated the anionic competitive special properties. Lastly, it examined the ability of As (V) elimination by the NZVI for actual samples from groundwater.

The major discoveries were the (HR-TEM) study of the original NZVI, which showed that ~90% of the nanoparticles were under 0.03 μ m in diameter and the structure was "core-shell." After three months, the XRD analysis showed that As (V) treated NZVI was steadily transformed into magnetite corrosion products. The reduction of As (V) to As (III) was proven using XPS; the removal was 25 % over three months. As (V) adsorption kinetics were rapid and occurred in several minutes following a first-order rate expression, with a removal efficiency of 100% and initial concentration of 1.0 mg/L using (0.1, 0.2, and 0.4 g/L) NZVI. Lastly, observing the impacts of competing anions has shown that HCO₃⁻, H₄SiO₄, and H₂PO₄²⁻ are probable meddling agents in the As (V) adsorption sequence.

4. Limitations of Arsenic Removal

In the arsenic adsorption treatment method, the major limitation is the influence of competing ions. In the nanoscale zerovalent iron (NZVI) approach, the impact of ions was investigated, and there were no adverse effects on

the As (V) uptake in the presence of (1 mM) concentrations of (HCO₃⁻, SO₄²⁻, and NO₃⁻) anions. However, both (H₄SiO₄⁰ and PO₄³⁻) and a concentration of (1 mM) decreased the uptake from 99.90% to 39.90% and 30.0%, respectively. Consequently, when the concentration was increased to 10 mM, the adsorption dropped significantly from 99% to 79% for the bicarbonate HCO₃⁻. Moreover, the uptake descended in the case of (10 mM) PO₄³⁻. However, it was reported that the same anions have fewer effects than observed in the As (V) case [40]. Furthermore, in the cupric oxide method, high concentrations of competing anions were investigated. The results showed that even with a high concentration of phosphates, cupric oxide could eliminate 83.0% of As (III) and 100% of As [30].

In the arsenic (V) removal technique by NZVI, the influence of the pH has been investigated where a 100% adsorption efficiency occurs at a pH range of (3-7) with the following condition As (V) (1 mg/L) on NZVI (100 mg/L). However, the adsorption efficiency dropped to 84.7% at pH 9.0 and 38 % at pH 11.0 [40]. It was also reported that the same behavior was observed in the As (III). Furthermore, the adsorption dependence on pH was observed when different iron oxides were used to remove arsenic (V). While in the biological treatment, the pH effect was not reported, knowing that the sample's initial pH was natural.

5. Conclusions

The impact of inorganic arsenic (III) and arsenic (V) on human health and the environment is significant. The conventional methods show many barriers to remove arsenic, and multiple steps are required to achieve the desired concentration. However, depending on the arsenic contamination site, the treatment method is more effective than other methods. For example, only filtration can decrease the arsenic concentration down to a certain extent in a location where iron is already present. Moreover, technologies like electrocoagulation may be used in such a place where electricity is easily available and inexpensive. The unconventional technologies discussed in this study demonstrated higher removal efficiency than the conventional techniques. However, the costs associated with these technologies are high; nonetheless, they are approved as eco-friendly concepts. Also, these technologies successfully removed arsenic using minimum steps and without adjusting any pH or oxidation state.

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