




Magnetite a potential candidate in the removal of poisonous arsenic from drinking water: a comprehensive review

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Abstract

A significant concern regarding human health is the toxic substances present in water. The existence of arsenic contamination, in potable water, has been acknowledged as a significant global issue pertaining to human health. Numerous countries, including the USA, India, Bangladesh, Canada, Japan, Poland, Hungary, etc., face this issue regarding the percentage of arsenic being more than the acceptable limit in potable water as per United State Environmental Protection Agency (USEPA), which is 10 µg/L. To get rid of this problem, various conventional technologies including precipitation, oxidation, adsorption etc., have been used, but unfortunately, most of these have been remained less effective due to their low efficiency to meet the requirement. In comparison to these traditional techniques, the other methods pertaining nanomaterials are found more promising due to available more surfaces to volume ration of the nanomaterials. In this regard, several nanomaterials have been studied; among these materials, magnetite (Fe₃O₄) is found as a potential candidate. The adsorbent Fe₃O₄ being magnetic in nature has great benefit from its separation point of view, since it can be isolated using an external magnetic field, while other nanomaterials face problems of separation after they are used as adsorbents. In order to realize the systems containing Fe₃O₄ nanomaterial as an adsorbent, it is essential to comprehend the entire process of arsenic adsorption by Fe₃O₄. Several matrices and surfaces have been used to coat the Fe₃O₄. In this review, at first, we aim to summarize the sources of arsenic, its harmful consequences on human health, and the technologies available for arsenic removal, which is accompanied by the detailed discussion of various methods and aspects of utilization of the Fe₃O₄ for removing arsenic from drinking water. In general, experimental conditions shows impact on efficiency; thus, this review also discusses the impact of various experimental conditions like pH, temperature, time, competing species concentration, adsorbent size, and doses on eradication of arsenic.

Keywords Arsenic removal · magnetite nanoparticles · adsorption · drinking water

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1 Introduction

During past several years, toxic elements in drinking water such as, aluminum, mercury, lead, chromium, and arsenic etc. due to the rapid industrialization of human society has been grown as a public health hazard [1–3]. The discharge of these contaminants in natural water threatens as well as aquatic life but also harmed the human health. Arsenic (As) is recognized as a hazard in humans and can often be found in contaminated water sources, including naturally occurring poor-quality water [4]. It is a element that enters the body gradually and causes symptoms to manifest 8–14 years after drinking water tainted with arsenic [5]. Although this period depends upon many factors such as amount and length of exposure, and varies from person to person depending upon their immunity level, ubiquitously, As element endures as oxides in water, soil, and sediments [6]. In nature, As present in 4 oxidation state viz., 0, 3, +3 and +5. The two types of As mainly found are (i) arsenite (H_3AsO_3 – As (III)) and (ii) arsenate (HAsO_4^{2-} – As (V)). In mildly reducing conditions As(III) exists as arsenious acid (H_2AsO_3^- , H_3AsO_3 , HAsO_3^{2-}), while As (V) is found as H_3AsO_4 , AsO_4^{3-} , H_2AsO_4^- , and HAsO_4^{2-} [7]. Arsenic compounds are always pH dependent [8]. As pH of the solution becomes lower than 9.2, As (III) occurs mostly as neutral H_3AsO_3 , whereas HAsO_4^{2-} and H_2AsO_4^- are major species of As(V). The pentavalent state As(V) is roughly 60 times less hazardous than the trivalent state As(III), primarily because As(V) tends to form divalent species in groundwater [9, 10]. As per WHO, the present status for the maximum contaminant level (MCL) for arsenic in potable water is 10 mg L^{-1} . The countries including Mexico, United State, Bangladesh, Vietnam, Romania and India are facing hazardous effects of arsenic-contaminated water [11]. As is introduced in drinking water through many ways such as, from rocks weathering, fertilizers, industrial waste, smelting of metals, volcanic emission, biological activities, agriculture application of pesticides and geochemical reactions [12, 13]. Although, efforts have been made to recent arsenic and its compounds production through environmental regulations but still its use in many industries, forestry and agriculture is a serious issue and needs an immediate attention. A variety of standard approaches are described for eliminating As from drinking, like adsorption, flocculation, ion-exchange, and electro-dialysis [14–17]. The conventional methods could remove arsenic from aqueous solutions but in actual practice their sorption capacities need to be improved for their practical applications. These limitations of conventional methods are overcome by researchers by using nanomaterials as novel substances to eliminate arsenic from water. In recent years, nanostructure adsorbents

have shown remarkable potential in order to address the environmental issues [18]. Application of nanomaterials in water refinement is found an effective way to remove the arsenic towing to large surface areas of nanomaterials. Nanomaterials can be more effective in water purification because of number of advantages such as adsorption of arsenic onto nanostructures materials has high efficiency and capacity materials because due to ease of operation and low cost [19]. Large number of nanomaterials such as, nano-metal/metal oxides, sorbents based of polymers, and carbon-based like, graphene, carbon nanotubes (CNTs), have studied to eradicate the poisonous arsenic from water, and exhibited maximum adsorption. Moreover, adsorption of both As from water has been explored by the mean of iron coated sand [20, 21], and granular ferric hydroxides [22]. In addition, minerals and ores as kaolinite [23], hematite and goethite [24], and feldspar [25] too have been in the focus of studies.

Among different materials, iron based materials have been found to exhibit significant adsorption affinity for arsenic compounds [26–28]. As the As(III) is identified extra venomous compare to As(V) [4], therefore, identification and adsorption studies of As(III) using different adsorbent such as, activated carbon, polymeric material coated with iron oxide sorbent etc. have been studied extensively [29, 30]. Studies revealed that the solid systems suggested above may have poor mass transportation with complex process, and therefore the dispersion of nanomaterials is one of the alternatives. Dispersed nanoparticles would have efficient mass transport to their surfaces and can be collected magnetically [31]. Furthermore, this approach eliminates numerous issues associated with filtration processes, such as blockages and fouling of packed membranes and columns. Thus, the magnetically separation using nanoscale magnetic materials is a better option and easy process.

Magnetite nanoparticles composed of Fe_3O_4 are promising candidates for adsorbing arsenic and have been recognized as a suitable option for purifying water contaminated with arsenic [20]. The magnetite nanoparticles tend to show stronger desorption hysteresis because as in our studies we found that the arsenic forms stable arsenic complexes which cover the surface of magnetite nanoparticles uniformly. Moreover, another advantage while using magnetite nanoparticles is their disposal at common environmental conditions.

In this review, we have presented a lot of aspects of magnetite nanoparticles for the purification of water. Initially, we have discussed the various available technologies for arsenic removal followed by synthesis and fictionalization of magnetite nanoparticles by different methods. The use of laboratory synthesized and commercially available magnetite nanoparticles in water purification in different research work has been discussed subsequently. Laboratory prepared

magnetite nanoparticles are compared with commercially available one [32], and the consequences of many factors, including size of nanoparticles, time, temperature, pH value of the solution; external species etc. on the arsenic removal efficiency are also presented successively.

2 Sources and occurrence of Arsenic

Several natural and human-induced factors contribute to arsenic impurity in groundwater sources. The As is rarely found in free state, mostly it combines with iron, Sulphur, and oxygen. Arsenic oxidation states depend upon its environmental conditions (mostly sulfides, metal arsenide's and/or arsenates. As(III) being of hard acidic nature formulate complexes with oxygen & nitrogen, while the As(V) being of soft acidic nature forms complexes with sulfides. Two major parameters that affect As speciation are pH and redox potential.

As occurs naturally in about 200 unique mineral forms, and in fact possesses about 60% as arsenates, 20% sulfides/sulfosalts, and 20% arsenite, arsenide's, oxides, silicates along with elemental As [33, 34]. Arsenopyrite (FeAsS), orpiment (As₂S₃) and realgar (As₄S₄) are the As containing minerals in which FeAsS mainly exist in the rock forming minerals and anaerobic conditions viz. sulfide, phosphate, oxide, silicate and carbonate, As₄S₄ and As₂S₃ are reduced forms of As and As₂O₃ is oxidized form of As [6, 35]. The major anthropogenic sources of groundwater contamination with As are mining, processing of various ores like Ni, Au, Pb, Cu and Zn, arsenical fungicides usage, fossil fuels burning, insecticides and herbicides in preservatives for woods and agriculture. Another source includes cotton and wool processing, seepages occurring through hazardous waste site, burning of arsenic coal contaminates with arsenic, and from the units manufacturing semiconductor and glass etc. [35, 36] However, these sources (anthropogenic) appear less responsible for contamination of groundwater than to the natural sources, although their role in groundwater contamination must be recognized.

First time as contamination was identified West Bengal and Bangladesh [37], Inner Mongolia and further areas of Asia were also reported to possess As contamination. The maximum contamination in these countries varies due to differences in sociopolitical and economic situations, as well as the lack of treatment technologies [38].

3 Hazardous effects of Arsenic

In the previous several years, several epidemiological and toxicological investigation have been done to determine the health concerns connected to ingesting arsenic. Animals and

plants may both be poisoned by arsenic, its toxicity effect decreases from arsine to elemental arsenic [39]. The inorganic As forms are determined to be more injurious compared to organic forms when taken by human beings, living organisms, and animals as they are proven carcinogens in humans [40]. Consumption of great amounts of As polluted water with lower exposure time results in acute and sub-acute poisoning while, its long-time consumption causes chronic poisoning. Arsenic cause poisoning by interfering enzyme action, essential cation, and transcriptional cellular events. Arsenic poisoning is categorized as Class 'A' human carcinogen by USEPA. Many researchers [41, 42] reported that the chronic As poisoning produces skin lesions, diabetes mellitus, hypertension, respiratory diseases, kidney, like lung cancer, liver, skin, and uterus.

4 Technologies and methods available for arsenic removal

As removal from water has been attempted via the variety of techniques, however the cost-effectiveness of the technologies makes them unaffordable on a broad scale. The technologies which are economically affordable are less effective for the treated water to attain the standards prescribed by World Health Organization, 1993. Following is a brief discussion of the existing technologies.

Khair, 1999 has found cellulosic material, bleached sawdust and pulped newspaper as the efficient materials in exclusion of As(III) as well as (V). These newspaper pulps coated by Iron (III) hydroxide are effective for arsenic adsorption at laboratory scale and in small scale home treatment units. Similarly, an institution working on allergy and skin care (AARSCI) has developed a simple cost filter beds using indigenous materials like coconut coir, husk, and shell with some amount of alum.

Arsen: X filter (a US company), has made a filter capable to remove arsenic as well as other contaminations such as fluoride and lead. Similarly, Project Earth Industries in USA has designed a unit to remove arsenic from the water pulled from hand pump tube well. This arsenic removal unit comprises of an adsorption media which adsorbs arsenic and other species present in the water. A company in Japan named Shin Nihon Co. Ltd., has made a house hold As elimination unit which is a plastic container in cylindrical shape that can be fitted with a tap at the bottom of water container used to outflow. To ensure optimal performance, the adsorbent material in this system is kept immersed. This technique is particularly good in removing arsenic from water that contains little iron. In South Korea, Coolmart Water Purifier has developed a purifier unit consisting a series of adsorbent beds having bio mineral sand, activated carbon, silica and zeolite

by which water is made to pass [43]. Although, lot of technologies are existing for As elimination but the best technology should be highly efficient in arsenic removal, easily achievable, geographical compatibility and applicability with the existing water treatment procedures.

4.1 Oxidation / Precipitation

As in water predominantly exists in 2 states: As(V) and As(III). The uncharged behavior of As(III) under 9.2 pH [44] effects most of the most treatment processes; however at the same time they are good in eliminating As(V). The soluble As(III) must therefore be changed into As(V), and As(V) must subsequently be precipitated. This process is generally required in the case, where As(III) exists predominantly at/near the neutral pH values.

It's been proved that the oxidation is a successful technique for As removal because As(V) gets readily adsorbed than As(III) onto solid surfaces [45, 46] As oxidation can be done by a number of oxidants such as, free available chlorine, ozone, MnO_4 , Cl_2O , FeO_4^{2-} , H_2O_2 , hypochlorite, potassium permanganate, Fenton's reagent ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$), MnO_2 coated nanostructured capsules etc. [26, 47–57] Moreover, many of the inorganic arsenic compounds are insoluble in water, which is the basis of arsenic removal technologies employing oxidation/ precipitation method [58]. Various oxidizing agents utilized by researchers for removal of arsenic and their removal efficiencies are mentioned in the Table 1.

This shows that the oxidation rate of As(III) is capable of being improved by exposing it to UV light with the oxygen presence, which has observed and reported numerously [53, 60]

The photocatalysis of As using TiO_2 was also explored in previous studies [63, 66]. When exposed to UV light, TiO_2 acts as both a photocatalyst and an adsorbent, but just as an adsorbent when not. The mechanism is demonstrated in Fig. 1.

Dodd et al. (2006) noted that the existence of natural organic material (NOM) enhances photochemical oxidation; however, at higher concentrations (2–15 mg/L), it decreases As(V) adsorption onto TiO_2 . Since NOM adsorption on TiO_2 influences the surface charges of the TiO_2 material, this drop in adsorption may be the result of competition within As(V) and NOM for accessible sites on the TiO_2 surface [52]. Therefore, by properly choosing the oxidants in respect to the water and chemistry compositions, the elimination efficiency of As can be increased. In addition, the process of oxidation alone removes a part of As and thus it is considered not to be an effective method.

4.2 Coagulation and Electro-coagulation

Coagulation is a widely recognized conventional method employed for As elimination from large amount of water by addition of chemical coagulants such as salts of Fe, Al, and Mn [68–71]. In the coagulation process, coagulate is formed by aggregation of fine particles in water. The As subsequently form precipitates with iron or aluminum ions present in the added coagulants, leading to their concentration. As is eliminated from the water by filtering the obtained coagulates. The coagulants are termed arsenic-borne coagulants. The coagulation using ferric cations was performed in the late 1960s century in the country Taiwan, where it was used to decontaminate deep water with elevated arsenic content [72]. Since then, a number of reports have been published using coagulation method for removing arsenic. This approach is found quite useful in eliminating both type of As. The pre-oxidation step in an important step to change the As. Song et al., (2006) examined coagulation for As elimination from water in a drainage system (38–74 μm) by employing ferric cations. The process removed more than 99 % of arsenic from a arsenic rich (5 mg/L) water. In this mechanism, where coarse calcite enhances coagulation in arsenic rich water, the double layer interface within coagulates and calcite is considered as one of the causing factors. The formation of double layer removes the potential energy barrier that exists between particles in a heterogeneous system [73].

Some studies have documented the usefulness of electrocoagulation (EC) for arsenic removal from water [74–76]. In the report via Kobya et al., (2011), arsenic was removed by the EC by performing a number of experiments with the variation of pH (4–9), current density in the experiment (1.75–7.5A/m²) and time of operation (0–15min) using iron and aluminum electrodes (in parallel and series). The elimination of 99.3% was achieved at monopolar series electrode connection mode for Fe at pH value of 6.5, and 98.9% was removed using Al electrodes at pH7. A comparative study carried out by Lakshmanan et al., (2010) for EC and chemical coagulation (CC) showed erratic removal of As(V) by EC at 6.5 pH. The observed higher removal in EC was due to the generation of Fe^{2+} . It is thought that a temporary rise in the pH in the case of EC process showed higher values of removal efficiencies. In this process; however, oxidation of As (III) could not be observed to a significant value and its adsorption on the iron hydroxides was nearly 5 to 30 % that of the As (V) [70]. In another study, Balasubramanian et al., (2009) conducted the arsenic removal experiments by electrocoagulation where they used steel sacrificial anode. Here, efficiency of 94% was achieved. Basically, in their studies they studied the effect pH of electrolyte on the As elimination efficiency as shown below [77, 78].

Table 1 Oxidants and corresponding removal efficiencies

Oxidants	Operating pH	Initial arsenic concentration	Type of water	Remarks	Reference
MnO ₂ coated nanostructure capsules	5.7-5.8	0.1-1.0 mg L ⁻¹	Laboratory water	Maximum oxidation percentages were 90% and 73% despite treating for 24h	[59]
<i>Photocatalytic oxidation</i> UV-C/H ₂ O ₂ and UV-A/ TiO ₂	7	500 µg L ⁻¹	ultrapure water (18.2 MΩ cm)	Peroxidation increases the removal percentage ; however, Fe(III) or Al(III) do not show any significant difference on the As removal	[60]
Iron based photo oxidation and sunlight assisted oxidation	2	0.47-4 mg L ⁻¹	Mine water and tube well water	100% oxidation. Sulphite concentration of 0.47 mg L ⁻¹ and 6 mg L ⁻¹ sulfite is required for highest efficiency	[56]
Solar and photochemical oxidation	6.5-8.0	500 µg L ⁻¹	Synthetic groundwater	There was more than 90% removal through 90 W m ⁻² UV-A irradiation for about 2-3 hours, using 0.06-5 mg L ⁻¹ of Fe(II,III) and 100 to 200 µL of citrate. The process resulted in oxidizing complex of iron-citrate	[58]
Solar light and iron citrate complex	-	2.09-5.58 µmol L ⁻¹	River and synthetic water	In about 4 h of irradiation, more than 90% of As was removed. The process showed faster removal of As(III) (80% removal in 1 h) than the As(V) (which took 4 h irradiation in 80% removal)	[61]
TiO ₂ photo-catalyzed oxidation	4.0-9.4	1-15 µM	Mili Q water	The study showed that As(III) oxidize fast when natural organic matter are present, even without using the oxygen and light	[62]
TiO ₂ photo-catalyzed oxidation	4.0-9.0	40-200 µM	Laboratory water	This study showed removal of As below 10 µg L ⁻¹	[63]
Photochemical oxidation in ferrioxalate solutions	3-7	1.3-13.500 µM	Laboratory water	In this process the maximum oxidation was achieved at pH 5	[64]
Photochemical oxidation assisted by peroxydisulfate ions	3	0.07-0.675 mM	Milli Q water	This study showed that the intensity of UV light dissolved oxygen are the important parameter in the oxidation of As(III)	[65]



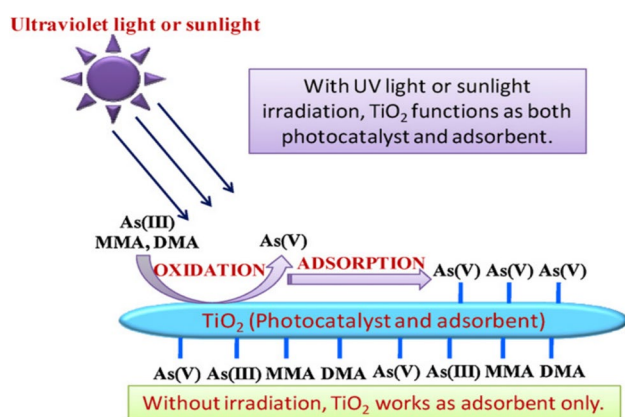


Fig. 1 Schematic representation showing TiO_2 for arsenic removal. Reprinted from [67]

4.3 Adsorption / Ion-exchange

In adsorption solid is used to eliminate impurities/substances from contaminated liquid solutions. The materials are separated after complete adsorption. This method has seen widespread application in As elimination from water. It is generally used in arsenic elimination, and it is reported that it reduces the arsenic contaminations less than $10 \mu\text{g L}^{-1}$; nevertheless, its performance depends upon the variety of water contaminants. In this process, as it is well known, the contaminated water is used to pass through an adsorbent filled column, during its passage the adsorbent surface adsorbs water contaminations and with the time the available sites are filled, after which the column needs to be refilled with the adsorbent material [79–85].

Gupta et al., (2005) executed batch studies and column studies for adsorption nature of As(III) via iron oxide-coated sand via varying the different parameters. The authors compared the results with those for uncoated sand. Adsorbent coated sand is reported to show a maximum efficiency of $28.57 \mu\text{g/g}$ as compared to that of uncoated sand $5.63 \mu\text{g/g}$ at pH 7.5 in 2 h adsorption for As(III). At 20 g/L adsorbent dose, adsorbent coated sand showed 99% removal of As(III) in batch study experiments [86]. In other studies, while using column studies, 94% efficiency was observed. Using iron–zirconium oxide adsorbent, Ren et al., (2011) found adsorption capacities of 46.10 mg/g for As(V) as well as 120.0 mg/g for As(III) at pH 7.0, which showed significantly higher adsorption capacity. For this adsorbent about 25 h took to reach the equilibrium for both contaminants. Presence of other anions hindered the adsorption in order $\text{PO}_4^{3-} > \text{SiO}_3^{2-} > \text{CO}_3^{2-} > \text{SO}_4^{2-}$ [87]. Kango et al., (2016) synthesized magnetite nanoparticles for the arsenic removal that were coated on the sand particles. Here magnetite nanoparticles act as an adsorbent. The authors performed batch experiment by altering arsenic content, adsorbent dose, and pH.

The synthesized adsorbent gives a noteworthy efficiency of approximately 99.6% for As(III), showcasing its remarkable arsenic removal capabilities. These synthesized adsorbent holds promise for addressing water filtration challenges in rural regions. In this study, simple filtration process can be used for nanoparticles extraction from water. Kinetic investigations indicated that equilibrium was attained within 360 minutes, aligning with the pseudo-second-order kinetics, which was more fitting compared to the pseudo-first-order kinetics [88].

In another effort by Tu et al., (2013) using copper ferrite which was formed from the electronic industry, using the scrap of printed circuit board the As(III) was removed. This substance suited the Langmuir isotherm well and had an L-shaped nonlinear isotherm, which indicated that there are limited binding sites, the adsorption occurs exclusively in the monolayer of the surface. The obtained maximum capacity for As removal was found 41.2 mg g^{-1} at a pH 4.2, which decreases while using the adsorbent at (pH > 9.2) [89].

Ion exchange, a unique kind of adsorption where there is no irreversible alteration to the solid's structure, it is the reversible exchange of ions within solid and liquid phase. While other forms of adsorption are less easily reversed as they involve stronger bonds. Usually, the solid resin forms a flexible three-dimensional hydrocarbon matrix containing multiple ionizable groups, electrostatically connected to resin. These groups are then replaced by ions in the solution with similar charges but higher exchange preference, or selectivity, for the resin. As eradication is done by passing water, with pressure, through one or more columns filled with resin. Notably, these resins demonstrate insensitivity to pH fluctuations within the pH range of 6.5 to 9.0 [90].

4.4 Lime softening

Lime softening works identically to metallic salt coagulation. When hydrolyzed lime (Ca(OH)_2) comes into contact with carbonic acid, it undergoes a reaction to produce calcium carbonate (CaCO_3). This CaCO_3 compound serves as an effective adsorbent for arsenic. This approach is typically employed only in case of very hard water with pH from 10 to 12 (Johnston and Heijnen, 2001). Between 9 and 10, on the pH scale a notable arsenic (As(V)) elimination of approximately 40–70% was achieved in water containing initial As amount ranging 0.1 to 20 mg/L. Subsequently, an enhancement in removal efficiency was observed through a sequential process involving lime softening, then coagulation with an iron salt. By implementing this method, a remarkable removal efficiency of 95% for As(V) was attained starting amount of 12 mg/L, with the optimal pH range being 10.6 to 11.4 [48, 91–93]. This method being costly is not suitable for small systems.

4.5 Membrane separation

Membrane processes have been known as a promising and diverse technology for effectively removing a wide range of toxins from water sources including arsenic, from water sources. This innovative approach holds the potential to remove As content to levels lower than 50 µg/L and, in certain instances, even less than 10 µg/L. Researchers have extensively explored four distinct membrane processes, namely microfiltration (MF), ultrafiltration (UF), reverse osmosis (RO), and nanofiltration (NF) as means to tackle As from dirty water. The efficiency of these procedures hinges upon the pore size of membranes. As the driving pressure increases, the selectivity of these membranes also rises, enabling them to effectively separate contaminants, including arsenic, from the water matrix (Siddiqui et al., 2017). Due of the low pressure nature of MF and UF, separation of contaminants is accomplished by mechanical sieving whereas NF and RO are high pressure processes, therefore separation takes place via chemical diffusion across the permeable membrane [7, 90, 94].

MF and UF membranes, operating under low-pressure conditions, demonstrate the capability to exclusively eliminate the particulate manifestation of arsenic from water. Conversely, the dissolved forms of arsenic remain unaffected due to their molecular sizes, allowing them to traverse through the membrane's pores. Consequently, the effectiveness of arsenic removal via MF and UF mechanisms hinges upon the composition of the As-bearing particles in water. As a result, to enhance the removal efficiency of these membranes, certain researchers have made use of these membranes with modifications for arsenic

removal. Han et al., (2002) utilized $\text{Fe}_2(\text{SO}_4)_3$ and FeCl_3 as flocculants in combination with microfiltration [95]

NF and RO processes, characterized by high-pressure conditions, exhibit the potential to drastically lower the amounts of dissolved arsenic in water. However, this efficacy is contingent upon the initial feed water containing minimal suspended solids [96]. According to Waypa et al., the utilization of both NF and RO technologies to eliminate As from freshwater and surface water sources was investigated. The results of the study demonstrated the impressive effectiveness of these membrane processes, as they achieved significant removal of both As, with elimination rates reaching 99% from the water samples. This observation implies that separation is caused by size exclusion rather than charge interaction [97]. Zhao et al. (2012) investigated As removal from aqueous solution that included As salts. The researchers employed a custom-made nanofiltration membrane composed of PMIA (poly m-phenylene isothalamate) and discovered that it achieved a remarkable arsenate rejection rate exceeding 90%. Significantly, they noticed an increase in arsenate (As(V)) exclusion from 83% to 99% with the pH elevation from 3 to 9 [98]. A compilation of arsenic species rejection percentages achieved by various membranes is provided in Table 2.

Reverse osmosis is the most popular and time-tested technique for eliminating arsenic from small water systems. Membranes featuring ultra-small pores, measuring less than 0.001 µm [111], find prominent use in water desalination applications, displaying impressive rejection rates of up to 99% for low molecular mass compounds [112]. However, it's worth noting that these membranes exhibit a significant drawback in arsenic removal—multiple studies indicate a higher rejection of As(V) related to As(III). Brandhuber

Table 2 Rejection (%) of arsenic by membrane. Reprinted permission from [99]

Membrane Used	As removal efficiency (%)	Reference
ZVI-Kaolin and PES	As (III): 50	[100]
Cu/CuO/poly(ethylene terephthalate) composites	As(III): 48.70	[101]
5% Zn/Al ₂ O ₃ /Polysulfone	As(V): >87	[102]
P-60S-EDTA membrane*	As(V): >96.75	[103]
PPSU/ZrO ₂ /CA hollow fiber membrane**	As(V): 87.24	[104]
PPSU/ZrO ₂ /CAP hollow fiber membrane**	As(V): 70.48	[104]
NF- flat sheet membrane	As(III) 67.72	[105]
RO-membrane	As(III) 94-99	[105]
Composite membrane with aromatic polyamide Selective layer	As(V): 40	[106]
NF-300 TFC polyamide	As(V): >95	[107]
NF90-4040 Polyamide TFC°	As(V): 94	[108]
NF90-4040 Polyamide TFC	As(III): 90	[108]
Dow/FilmTec NF90 Polyamide	As(V): 98	[109]
TFN°	As(V): 98.6	[110]
NF-90 membrane	As(V): 98.4	[96]

et al.,(1998) performed arsenic removal experiments employing four different RO membranes. Their findings indicated that all RO membranes rejected 95% of As (V) and approximately 65-85% of As (III). The literature also showcases instances of standalone arsenic treatment using reverse osmosis (RO) units, attaining over 97% elimination of As(V) along with more than 92% of As(III) [113]. A summary of arsenic removal achieved by select commercial reverse osmosis membranes is given in Table 3.

4.6 Foam floatation

Low As content can be eliminated by Adsorbing Colloid Floatation (ACF) [122, 123]. ACF is a technique that entails introducing a coagulant, such as ferric hydroxide, alum, iron (III) sulfate, or goethite, to generate aggregates known as flocs. Within this process, arsenic becomes adsorbed onto these flocs and undergoes co-precipitation alongside them. Subsequently, the application of a surfactant facilitates the collection of these flocs. ACF is recognized as an analytical approach designed to concurrently eliminate boric acid, arsenic, and germanium from a solution [124, 125]. This efficacy is detected within a pH 4 to 5 [126]. But this technique is not selective for arsenic.

4.7 Bioremediation

Biological approaches encompass two main categories: phytoremediation, which employs plants to remove or stabilize elements, and biological treatments that utilize microorganisms. Phytoremediation methods involve the use of specific plant species such as sunflower, Indian mustard, poplar, maize, cottonwood, and various grasses (such as prairie and ryegrass). These plants carry out either Phyto stabilization or phytoextraction to either stabilize or remove arsenic from groundwater. Additionally, there are hyper-accumulating ferns that have the unique aptitude to accumulate huge concentrations of As within their above-ground tissues [127].

The Chinese brake fern *Pteris vittata* is the first known As hyperaccumulator. It can accumulate arsenic at levels ranging from 12 to 64 mg/kg from soils with natural arsenic concentrations of 0.5 to 7.5 mg/kg, and even up to an impressive 22,630 mg/kg when grown in soils amended with 1500 mg/kg of arsenic [128]. Other fern species, such as *Pteris cretica*, *Pityrogramma calomelanos*, *Agrostis stolonifera*, *Pteris umbrosa*, *Pteris longifolia*, and *Agrostis tenuis*, have also demonstrated the ability to hyperaccumulate arsenic [127].

The occurrence of As hyperaccumulation in plants forms the basis for As phytoremediation techniques. When it comes to groundwater cleanup, plants with extensive root

Table 3 Rejection (%) of arsenic by RO membrane. Reprinted permission from [114]

Removal Process	Experiment Conditions					Removal Efficiency %	Ref.
	Membrane Type	Concentration	Temperature	Pressure	pH		
Nanofiltration	NF-300	100–382 µg/L	10–25 °C	310–724 kPa	1.2–8.8	As ⁵⁺ : 95	[115]
Nanofiltration	NF90-2540	100–1000 µg/L	27–37 °C	4–7 bar	3–11	As ³⁺ : 86.75–95.11 As ⁵⁺ : 94.13–99.02	[116]
Nanofiltration	NF90	20–100 µg/L	25 °C	138–552 kPa	4–10	As ³⁺ : 44 As ⁵⁺ :89	[117]
Nanofiltration	NF30	100–1000 µg/L	15–40 °C	2–12 bar	3.1–5	As ⁵⁺ : 77-88	[96]
Nanofiltration	NF90					As ⁵⁺ : 94.4–98	
Nanofiltration	NF	50–400 µg/L	20 °C	0.41–0.82 MPa	2–10	As ³⁺ : 10-40	[106]
Reverse osmosis	LPRO	-	-	-	-	As ³⁺ : 65-90	
Nanofiltration	NF90	100 µg/L	20 °C	3.1–5 bar	3.1–5	As ³⁺ : 67.72	[105]
Nanofiltration	NF270	-	-	-	-	As ³⁺ : 57.96	
Reverse osmosis	XLE					As ³⁺ : 98.23	
Reverse osmosis	BW30					As ³⁺ : 97.47	
Reverse osmosis	PVD	69.3 µg/L	8–21 °C	5–14 bar	5–9	As ³⁺ : 98.1	[118]
Reverse osmosis	TFC-SR					As ³⁺ : 96.1	
Reverse osmosis	FT30					As ³⁺ : 89.2	
Reverse osmosis	TFC-ULP					As ³⁺ : 83.2	
Reverse osmosis	BW30					As ³⁺ : 90	
Reverse osmosis	TE2521	0.0–2.5 mg/L	25–30 °C	190-210 psi	6–8	Total As: 95-99	[119]
Reverse osmosis	SWHR	50–750 µg/L	20 °C	10–35 bar	4.1–9.1	As ³⁺ : 92.5 As ⁵⁺ : 96.8	[120]
Reverse osmosis	TW40	0.2–18 mg/L	4–30 °C	190 bar	5–9	Total As: 95-99	[121]

systems are preferred for their enhanced capability to reach arsenic-contaminated layers more efficiently [129]

Giri and Patel (2012) conducted a study employing the aquatic plant *Eichhornia crassipes* to assess the phytoaccumulation effectiveness of total arsenic (As). During the experiment, the plants were cultivated in double-distilled water along with a customized 0.25 N Hoagland's nutrient at pH 6.8. Various amounts of arsenic (III) and arsenic (V) were supplemented in equal proportions of 0, 0.010, 0.025, 0.05, and 0.10 mg/L. Harvesting of plants were done at intervals of 0, 3, 9, and 15 days. The results indicated that plants treated with a solution containing 0.10 mg/L of As, accrued the highest concentrations in their roots (7.2 mg/kg, dry weight) as well as shoots (32.1 mg/kg, dry weight) after a cultivation period of 15 days. Similarly, biological treatments have been applied in many innovative techniques for the arsenic removal from water [130–133].

4.8 Solvent extraction

The relative solubility of substances in two separated immiscible liquids, including water and an organic solvent, is what determines how easily a material may be extracted from one liquid phase to another. This method has been used by many researchers for water purification [134–136] have found that the extraction efficacy is affected by factors such as the choice of diluents, extractant, and the valency of arsenic present in water. Notably, aromatic diluents, like toluene, have been identified as favorable options for diluents in the context of arsenic removal [137]. In this method, complete arsenic removal and disposal of byproducts are also the main problems.

4.9 Reactive barrier

One way to remove arsenic is to employ iron in Permeable Reactive Barriers (PRB) [138, 139]. PRB is basically a reactive material zone in an arsenic removing system. A conventional design for PRBs involves creating a continuous trench through excavation and backfilling with a reactive substance. The selection of reactive material, for example, activated carbon, limestone, iron, zeolites, or compost, depends on the specific contaminants targeted for remediation. This approach employs a "funnel and gate" system, which guides contaminated groundwater using low-permeability walls toward a permeable treatment zone. In many practical PRB systems, especially those addressing issues related to chlorinated organic compounds, metals, and radionuclides, zero-valent iron is an often-used media. This material acts as a reducing agent, aiding in the conversion of dangerous pollutants into less toxic or immobile forms. Consequently, the PRB functions as a barrier intercepting and treating contaminated groundwater, thereby reducing contaminant

Table 4 Comparison of competence for removal of arsenic by several treatment techniques and wastage of water Reprinted permission from [141]

Method Employed for Purification	Competence for Removal of Arsenic (in %)	Wastage of water (in %)
Oxidation and Filtration		
Greensand	50-90	<2
Biological Oxidation	>95	<2
Co-precipitation		
Enhanced Lime Softening	90	1-2
Enhanced Coagulation/Filtration	<90	1-2
With Alum	95	1-2
With Ferric Chloride	90	5
Coagulation Assisted Microfiltration		
Adsorption	95	1-2
Activated Alumina		
Iron based sorbents	Up to 98	1-2
Ion exchange	95	1-2
Membrane Technology		
Reverse Osmosis	>95	15-20

concentrations before they can affect the surrounding environment downstream [140].

4.10 Comparison of several arsenic removal techniques

Each of the discussed methods for arsenic removal comes with its own set of benefits and drawbacks. Table 4 lists the water losses and arsenic removal efficiency for several technologies when used in typical circumstances. Adsorption is the most efficient approach out of all of them due to the inexpensive nature of treatment, large removal efficiency, and minimal water loss. Among various sorbents, iron-based sorbents, including magnetite and maghemite, have high arsenic removal efficiency (up to 98%) with 1-2% water loss only [142]. Thus, in the present review authors discussed about arsenic removal using magnetite nanoparticles (iron-based sorbents) as adsorbents.

5 Magnetite nanoparticles

Magnetic nanoparticles are widely employed in water treatment because they are simple to find and utilize again. Magnetite, a compound composed of Fe(II) and Fe(III) salts, with unique superparamagnetic characteristics, accessibility, environmental friendliness, high saturation field, chemical stability, low Curie temperature, non-toxicity, and absorption capabilities [143]. It is commonly found in iron ores,

making it a cost-effective and sustainable option for catalysis, given iron's abundance among transition metals. Magnetite has found extensive use in eliminating contaminants from various water resources, employing both monometallic iron nanoparticles and bimetallic iron alloys or core-shell nanoparticles. Magnetite can be used as a reusable heterogeneous catalyst to transform contaminants into useful products in addition to eliminating pollutants. Its regulated size and shape, non-toxic traits, relative inertness, and magnetic nature make it a desirable catalyst support [144]. In recent past, the magnetite nanoparticles have been fabricated by the researchers and academicians by employing numerous techniques like sol-gel, hydrothermal and thermal decomposition [145], electrochemical, and sonochemical approaches, have been established for preparing magnetic nanoparticles (MNPs). A list of some commonly practiced fabrication techniques for this purpose along with their references has been given in Table 5.

Magnetite is a very promising material and shows a great potentiality about elimination of heavy toxic metals from water. For targeting this application of magnetite, some requisite modifications which are applied on the nanoparticles surface plays a pivotal role to enhance the stability of these particles inside their aqueous solutions as well as to increase the value of their exchange affinity. In the recent past, by incorporating some necessary changes in the prevalent fabrication methods, these requisite modifications on magnetite surface can be realized by their functionalization with some inorganic and organic materials. The modification of magnetite by coating them with silver, gold and silica not only helps to improve their aqueous stability, but it also helps to create suitable sites on their surface for the attachment of some ligands through covalent bonding as reported in the literature [153–155]. The researchers have also successfully developed numerous techniques to produce magnetite nanoparticles which are dispersible in their aqueous solutions to a great extent and exhibit superparamagnetic behavior as well. Besides all this, the size of these particles can be easily controlled and their surfaces can be further modified to show some

properties which are beneficial in water purification process. Chang et al., (2009) conducted synthesis involving the incorporation of aluminosilicate into magnetite, resulting in formation of nanocomposites. By utilizing the method of chemical precipitation, they first synthesized magnetite nanoparticles. Thereafter, by practicing sol-gel or acid treatment method, they deposited a thin layer of silica on magnetite surface and finally, by following sol-gel method, they developed aluminosilicates [156].

Cao et al., (2012) decomposed iron (III) acetylacetonate thermally in presence of methoxy polyethylene glycol, a multifunctional organic material that not only acted as a solvent, but as reducing agent as well in this decomposition reaction. In addition to this, by following a literature described procedure, they also fabricated MNP_s by decomposing Fe(acac)₃ thermally in presence of PEG (PEG-1000, Mw = 1000, 99%) for the sake of comparison [157, 158].

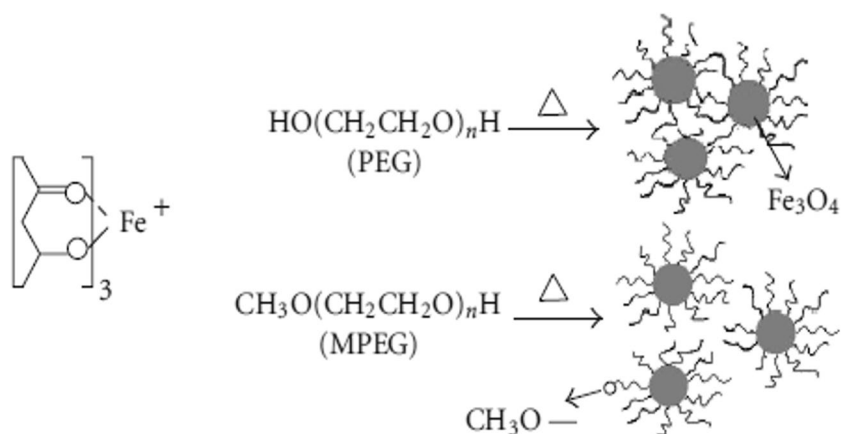
In case of MNPs coated with MPEG, due to the presence of some inert groups at their terminals, the network structural formation is not observed; whereas in case of MNPs coated with PEG the network structural formation is readily seen (Fig. 2).

Xin et al., (2012) synthesized magnetite nanoparticles showing mesoporosity which were modified on the surface with amino-group by following a new environment friendly procedure. They performed ninhydrin test to ascertain the amount of amino-group that got grafted onto surface of magnetite (AF-Fe₃O₄). At pH 7.0 each removal of 50 mL 5 mg/L of Pb(II), Cu(II), and Cd(II) on 10 mg of surface modified magnetite nanoparticles with amino-group attained equilibrium in 2h [159]. Tan et al., (2008) successfully reduced iron (III) acetylacetonate (Fe(acac)₃) with pyrrole or N-methylpyrrole by using oleylamine and oleic acid as surface active agents in air to fabricate magnetite nanoparticles having cubic and faceted uniformly dispersed structure [160]. Jing et al., (2012) by making use of sodium citrate and following a reversibly precipitating chemical reaction based simplified single-step method, they got success in producing magnetite (Fe₃O₄) nanoparticles whose surface was functionalized with carboxylate. These environment friendly particles were

Table 5 Different methods employed for fabrication of magnetite nanoparticles

Sr. No.	Synthesis methods	Composition	Arsenic removal	Reaction condition		References
				Adsorbent Dose	pH	
1.	Co-precipitation	Fe ₃ O ₄	95%	4 g/L	5.5	[146, 147]
		M-FeHT	95%	5 g/L	9.0	
2.	Sol-gel Synthesis	FS	38.2 mg g ⁻¹	0.15 g/L	7.0	[148, 149]
		γ-Fe ₂ O ₃	25 mg/g	100 mg/L	3	
3.	Co-precipitation, Hydrothermal	Fe ₃ O ₄ , Fe ₃ O ₄	~97%	81.04 mg g ⁻¹	2-6, 2.5	[150, 151]
4.	One pot synthesis	Fe ₃ O ₄	99.2%	10 μg/L	6.1	[11]
5.	Chemical reaction	Fe ₃ O ₄ -rGO	99.9%	0.2 g/L	7	[152]

Fig. 2 Schematic representation of decomposing $\text{Fe}(\text{acac})_3$ thermally in MPEG and PEG presence for fabricating magnetite nanoparticles. Reprinted from (Cao et al., 2012), open access 2012 [157]



easily dispersible in their aqueous solution and exhibited superparamagnetic character as well [161].

Saiz et al., (2014) first by utilizing co-precipitation method produced magnetite nanoparticles and thereafter through hydrolysis and condensation of tetraethyl orthosilicate got success in depositing a layer of mesoporous silica onto their surface. As synthesized aminopropyl-functional particles entrapping Fe^{3+} (S1-F3) inside them were further examined with regard to their adsorption efficiency in respect of underground water containing As^{3+} and As^{5+} ions

[162]. Navarathna et al. (2019) developed Fe_3O_4 /Douglas fir biochar composites (MBC) using a modified approach. The SEM image in Fig. 3 reveals morphological changes attributed to the widespread distribution of iron oxide precipitation. This precipitation is observed as primary Fe_3O_4 particles with a diameter of 18.1 ± 3.9 nm, often clustered to sizes ranging from 2 to 7 μm . The particle size was measured to be approximately 1650×1334 nm^2 . SEM-EDX analysis showed decreasing surface carbon going from BC to MBC because of the presence of Fe_3O_4 [163].

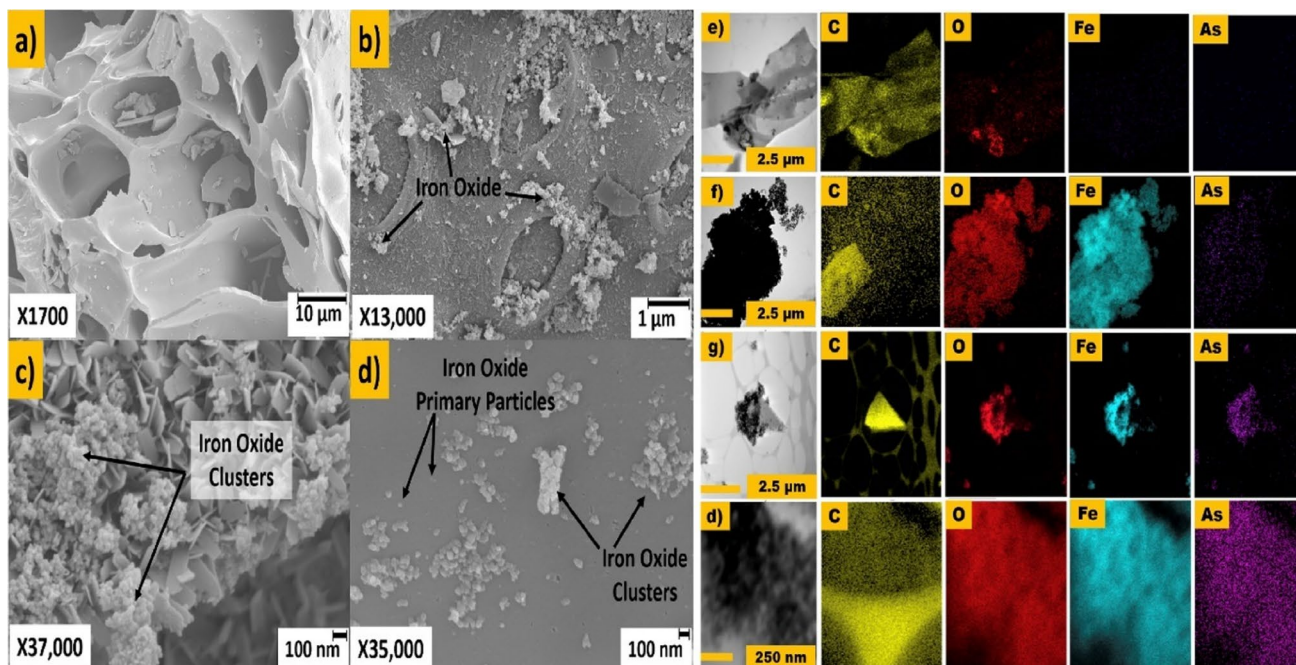


Fig. 3 SEM of BC (a) and MBC (b–d) and TEM and TEM-EDX elemental mapping of (e) BC (f) MBC (g) As-loaded MBC (h) As-loaded MBC [163]

6 Role of magnetite nanoparticles in adsorbing arsenic from polluted water

6.1 Laboratory synthesized magnetite nanoparticles and arsenic removal

There have already been a lot of studies done on creating magnetite nanoparticles in the lab and using them to eradicate As from polluted water. Ohe et al., (2010) investigated the adsorptive properties of laboratory synthesized high specific surface area magnetite nanoparticles at a temperature of 303 K for removing As(V) as well as As(III) ions. The adsorption abilities were found to be $2.77 \times 10^{-1} \text{ mmol}\cdot\text{g}^{-1}$ and $2.28 \times 10^{-1} \text{ mmol}\cdot\text{g}^{-1}$ for As(III) and As(V). The reported values of $4.74 \text{ dm}^3\cdot\text{mmol}^{-1}$ and $12.1 \text{ dm}^3\cdot\text{mmol}^{-1}$ adsorption equilibrium constants for As(III) and As(V). Indicates the higher affinity of these particles to As(V) [164]. D’Couto et al., (2008) used laboratory synthesized surfactant free magnetite nanoparticles for arsenic removal. The authors have used a column filter containing a layer of synthesized magnetite nanoparticles, and achieved more than 95% arsenic removal from a 100 ppb contamination level [165]. Yoon et al. (2017) developed magnetite/non-oxidative graphene (M-nOG) for eradication of As. While their study showed lower efficacy of M-nOG for eradicating As compared to graphene-based composites like magnetite-graphene oxide, it demonstrated notably higher efficiency compared to other magnetite composite adsorbents. In Fig. 4, it is depicted that adsorption initially inclined and then leveled off with increasing As content due to the saturation of available sites. At 25°C and pH 7, M-nOG exhibited highest adsorption of 14 mg/g for As(V) and 38 mg/g for As(III) [166].

Khodabakhshi et al., (2011) showed the role and the efficacy of newly synthesized magnetite nanoparticles for adsorption of As (III) present in industrially produced artificial water and found considerable potential of magnetite nanoparticles in As(III) removal. The authors carried out a detailed adsorption related studies for magnetite nanoparticles and consequently found that the data was in close agreement with Freundlich isotherm equations. The capacity of magnetite nanoparticles at pH=7 for adsorbing As (III) ions was found to be equal to 23.8 mg/g [167].

Parsons et al. (2009) showed possibility of recently synthesized magnetite nanoparticles with grain sizes of 28 nm for As(III) and As(V) eradication from aqueous solutions. These laboratories’ synthesized magnetite nanoparticles showed binding efficiency of 32.2 $\mu\text{g}/\text{g}$ for As(III) and 1575 $\mu\text{g}/\text{g}$ for As(V), respectively [151]. Singh et al., (2011) used laboratory synthesized nanoparticles of magnetite after duly functionalizing them with different groups like thiol (2,3-dimercaptosuccinic acid), amine (ethylenediamine) and carboxyl (succinic acid). The as synthesized particles were then tested on toxic ions like As(III), Cr(III), Cd(II), Pb(II), Ni(II), Co(II), and Cu(II) for their removal from polluted water samples [168]. Stefusova et al., (2012) observed co-precipitation of Fe(III) as well as Fe(II), which enabled them to successfully fabricate magnetite nanoparticles under an alkaline condition. As a result, they concluded that these laboratory synthesized nanoparticles can show a highest value of about 46.7 mg/g so far as their ability of sorption at a pH value of 3.5 is concerned. Besides all this the authors attempted the removal of arsenic from preloaded particles of magnetite under different states [169].

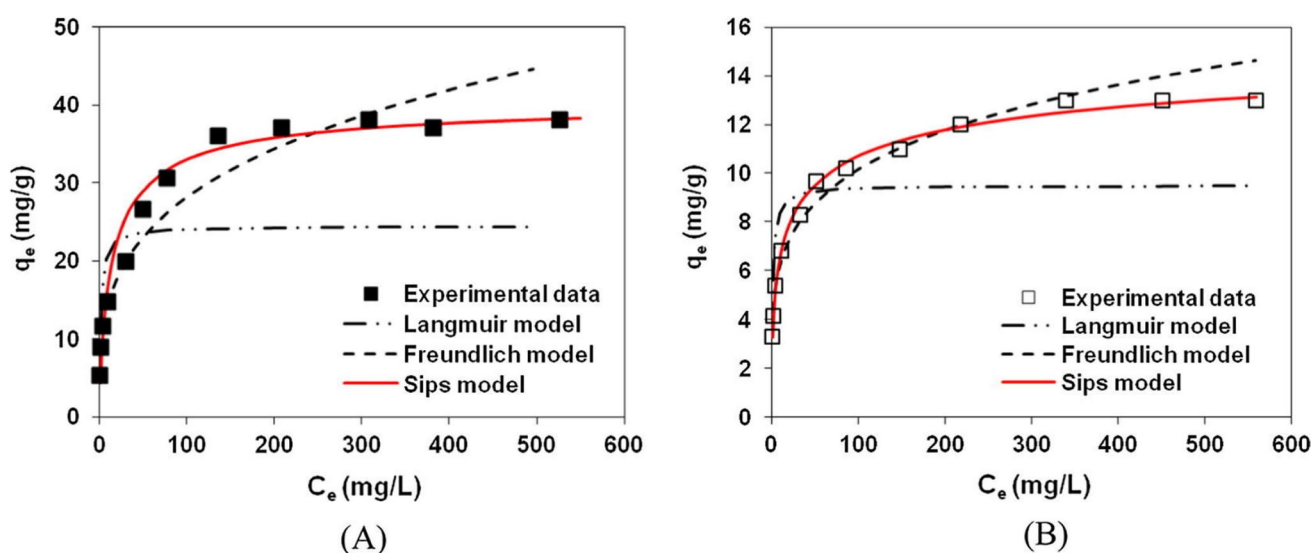


Fig. 4 Adsorption isotherms of As (A) As(III), (B) As(V) (M-nOG). Reprinted permission from [166]

6.2 Commercially available magnetite nanoparticles and arsenic removal

In recent past, Arsenic adsorption efficiency of mercantile nanoparticles of magnetite has been investigated via researchers extensively. Turk et al., (2010) purchased magnetite nanoparticles of less than 50 nm size from Sigma Aldrich and used them as an adsorbing material for removing As(V) from polluted water samples in varied conditions. They completed the study by the variation in the amount of metal ions (100 to 2,000 $\mu\text{g/L}$) initially, the time of contact (15 to 180 min.), pH value of solution (3–12) and the dose of adsorbing material (0.05–5 $\mu\text{g/L}$). Authors also performed some experimental studies in batches to investigate the detailed kinetics involved in As(V) adsorption and isotherms related to this study. The data pertaining to all these experimental studies was found most suitably justified by the pseudo-second-order kinetic theory. The adsorption of arsenic as obtained from equilibrium experiments was fitted by using Freundlich and Langmuir isotherms. Nano magnetite was found to show high removal efficiency for arsenate as it reduced the initial level from 300 to less than 5 $\mu\text{g/L}$, i.e., far lower than the optimum value (10 $\mu\text{g/L}$ of As) advised for drinking water by WHO [170].

Shiple et al., (2018) carried out arsenic adsorption experiments on mercantile nanoparticles of magnetite of the size of the order of 19.3 nm through column studies. Here they used 1.5 and 15% nanoparticles of magnetite by their weight and Lula soil in a column and passed a solution of arsenic formed with water inside it. For removing the both type of As) from a sample holding 100 $\mu\text{g/L}$ of them initially, 1.5 % of nanoparticles of magnetite by their weight were flowed at 1.5 and 6 mL per hour rate. Here arsenic was found to be completely freed at 400 PV. Authors too performed a long duration experiment with 15% nanoparticles of magnetite by their weight which were flown at 0.3 mL per hour rate and taking 100 $\mu\text{g/L}$ of As(V) initially. After a total volume of 3559.6 pore volumes (PV), which corresponds to approximately 132.1 days, no amount of As(V) was found but its amount was found to show 20% value after 9884.1 PV (207.9 days). The authors also enumerated a deceleration factor of around 6742 and that revealed a strong affinity of nanoparticles of magnetite to arsenic ions inside the column. It is also observed here that this affinity of nanoparticles to arsenic ions got enhanced on interrupting the flow of the solution. Chowdhury and Yanful (2010) used mercantile nanoparticles of magnetite of 20 nm size for removing arsenic from artificial as well as naturally occurring underground water. At a pH value of 2 and initially starting with an amount of As(III) equal to 3.69 mg/g and of As(V) equal to 3.71 mg/g, the maximum adsorption was found to 1.5 mg/L for these two ions in case of synthetic water. Under room temperature conditions, the nanoparticles of magnetite

demonstrated an adsorption capacity for arsenic amounting to 62.66 mmol/g, as given via Langmuir isotherm model. It further indicated that the nanoparticles of magnetite can spontaneously adsorb arsenic. It is the electrostatic force of attraction between magnetite nanoparticles and arsenic, which has been held responsible for elimination of As from polluted water samples in this study. Chowdhury et al., (2010) also showed the As removal efficiency of commercially available maghemite -magnetite- mixture with 20 to 40 nm particle size from aqueous solution for different pH values. They have achieved 96-99% arsenic removal under controlled pH conditions [171].

6.3 Comparative studies of laboratory synthesized and commercially available magnetite nanoparticles in arsenic removal

The literature contains very little information regarding comparative studies of As removal using laboratory prepared and commercially available magnetite nanoparticles. Based on numerous studies, this can be concluded that the efficiency of these particles to remove arsenic is basically determined by the nanoparticles' size. Mayo et al., (2007) made 12 nm size nanoparticles by mixing FeO(OH) (2.00mM), 1-octadecane (200mM) and oleic acid (8.00mM) and heating it at 320°C for a particular time period. Thereafter, they studied the impact of size of magnetite nanoparticles on their efficiency to remove and release As(III) as well as (V). Further it was observed that commercially available 20 nm particles had been found irreversibly captured inside the column, whereas the laboratory-synthesized nanoparticles were reversibly captured by the column and could be released back by it [11]. Similarly, Yean et al., (2005) have found a similar capability for adsorbing As(III) as well as (V) by the commercially available magnetic nanoparticles. The adsorption is found significantly increased for laboratory prepared 11.7 nm nanoparticles due to the more adsorption sites exposed for arsenic. It was found that small size magnetite nanoparticles have about 200 (wt %) times arsenic removal capacity, and show stronger desorption hysteresis, presumably synthesis of arsenic-iron compounds with high stability invariantly over the entire nanoparticle surface. Moreover, smaller nanoparticles oppose desorption of arsenic, therefore, a recovery mechanism based on application of small magnetic field but having high efficiency can be formed by carefully optimizing the particle size for water purification.

6.4 Magnetite nanoparticles in different complex forms for arsenic removal

The usage of various complexes of magnetite nanoparticles for As removal from water samples has previously been thoroughly investigated by the researchers. Activated carbon

fiber (ACF) doped with nanoparticles of magnetite was capable to decrease the As(V) level underneath the MCL recommended value [172]. Fe^{2+} and Fe^{3+} ions in a solution of ammonia with water are utilized to produce nanosized magnetite particles. ACF doped with magnetite nanoparticles were prepared by mixing chitosan solution (solution of 0.01 glacial acetic acid mixed with chitosan powders) with solution of polyethylene glycol (PEG), and then magnetite suspension was prepared. Resulting mixture was then applied on activated carbon fiber. After removing the excess mixtures by vacuum filtration, activated carbon fiber cloths were dried in an oven at 60°C followed by the washing. Finally, ACF doped with magnetite nanoparticles was heated in a furnace at 102°C for suitable time span (nearly 2 hours). Thus modified ACF clothes having an amount of 0.7g/L were capable to decrease As(V) below $10\mu\text{g/L}$. While raw ACF clothes had almost no capability to adsorb an amount of As(V) below $100\mu\text{g/L}$. Since As content in drinking water is about tens of $\mu\text{g/L}$, therefore, modified ACF can be a competent adsorbing material for removing arsenic from polluted water samples. Moreover, arsenic concentration in the natural water typically falls within a pH range of 5–8, and modified ACF exhibit an extended pH range spanning from 2–8, which is wider compared to raw ACF (pH = 4 to 6.8) [173].

Magnetite nanoparticles decorated multiwall carbon nanotubes (MWCNTs) super capacitor has been prepared for arsenic removal [174]. MWCNTs were fabricated via chemical vapor deposition method and were coated with magnetite nanoparticles after purification and functionalization in order to get a super capacitor. Chemical method using aqueous solutions of FeCl_3 , FeSO_4 , and NH_4OH was used for the coating purpose. The electrochemical performance of the synthesized nanoparticles concerning As in water demonstrates an adsorption efficiency of approximately 58% for As(V) and 67% for As(III) over 15 repeated cycles. This efficiency was achieved using 100 mg of nanocomposite loading on each electrode. This capacitor with nanocomposite on its plates is capable for arsenic removal by repeating the cycles of purification.

Magnetically active hybrid sorbent beads can remove As(V) at normal pH values. By employing a typical procedure, this kind of hybrid sorbent beads (MAHS) can be synthesized by precipitating nanoparticles of magnetite within an adsorbing polymer [175–178]. Experimentally, it has been proven that these beads are efficient to adsorb arsenic and chlorine - organic compounds altogether from drinking water [142].

A composite comprising magnetite particles (10 nm), combined with reduced graphene oxide, exhibits superparamagnetic properties at ambient conditions and can be extracted via magnetic field [152]. This composite exhibits an affinity for As(III) & (V), and 99.9% adsorption efficiency for arsenic at 1 ppb in comparison to plane nanoparticles

of magnetite. The as formed composite material is highly practicable for separation of arsenic from H_2O , reason being its strong affinity to arsenic because the production of many adsorption sites on magnetite-reduced graphene oxide composite.

Zirconium-based adsorbent $(\text{ZrO}(\text{OH})_2 \cdot 1.6\text{Fe}_3\text{O}_4 \cdot 2.5\text{H}_2\text{O})$ was developed using co-precipitation method by [179]. A solution of $\text{Zr}(\text{SO}_4)_2$ and $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ was made at a particular temperature which was followed by the addition of NaOH. For these magnetic nanoparticles, the majority of As(V) absorption happens fast in 8 h, which then becomes slow and attained equilibrium in 25 hours. The best absorption was seen at pH of 2.6 to 3.3, and it dropped significantly as pH increased. The presence of this adsorbent in As(V) solution at various starting pH levels causes a buffering effect due to the appearance of amphoteric surface hydroxide sites on the metal oxide.

The replacement of Fe^{2+} from magnetite nanoparticles by Mn^{2+} increases the adsorption capacity of arsenic [151]. MnFe_2O_4 nanoparticles can be produced via co-precipitation using FeCl_2 , MnSO_4 and NaOH solutions. Adsorption of As(III) and As(V) to the MnFe_2O_4 nanoparticles is almost independent to the pH values, except a slight increase for pH greater than 2. Similarly, in other study, MnFe_2O_4 , CoFe_2O_4 and magnetite nanoparticles were compared [180]. The highest adsorption of As(III) on MnFe_2O_4 and CoFe_2O_4 were determined to be 94 and 100 mg/g, while for As(V), they were found to be 90 and 74 mg/g, within a pH range of 3–10. It was about two times higher than those of magnetite nanoparticles (50 mg/g and 44 mg/g for As (III) and (V)), and were independent to the surface area. Therefore, surface properties, especially hydroxyl group present on metal surface (M–OH), are supposed to influence elimination of As from water. According to the mechanism, substitution of Fe^{2+} with Co^{2+} and Mn^{2+} leads to a remarkable enhancement in the amount of metal hydroxides (M–OH) inside this kind of nanomaterials. Arsenic removal was possible through substitution of these hydroxyl groups with As(V) as well as (III) to make singly-coordinated also doubly-coordinated metal complexes. Similarly, Ni and Co substitution in the $\text{Fe}_{1-x}\text{Ni}_y\text{Fe}_2\text{O}_4$ and $\text{Fe}_{1-x}\text{Co}_x\text{Fe}_2\text{O}_4$ ($x, y = 0, 0.05, 0.1, 0.2, 0.5$) nano-sized particles results in an effective arsenic adsorption for pH less than 7, which decreases further at higher pH values [181].

To eliminate both As(III) and As(V), hematite and oleic acid-coated magnetite nanoparticles present a viable option. These coated magnetite nanoparticles can be synthesized using the ball milling technique [182]. The minimum added quantity of hematite coated magnetite nanoparticles was about 100 mg per L for As(III) and 60 mg per L for As(V) to remove arsenic efficiently, which can reduce the original arsenic amount from $100\mu\text{g/L}$ to less than $10\mu\text{g/L}$. Furthermore, the recovery of hematite coated magnetite

nanoparticles is pH dependent. Whereas in case of oleic acid coated nanoparticles of magnetite the amount of arsenic can be decreased to a value as low as 45 µg/L and they are also capable of protecting the characteristics that are related to their magnetic behavior at separate pH values. The shielding done by surfactant molecules on the nanoparticles surface plays most crucial role in creating the difference in adsorption capacities of these adsorbents.

7 Factors influencing arsenic adsorption capacities

7.1 Impact of phosphate

Phosphate can form three kinds of compounds with magnetite on its surfaces (*i.e.*, goethite, hematite, etc.) *viz.*, with hydrogen ion ((FeO)₂(OH)PO), doubly coordinated without hydrogen ion ((FeO)₂PO₂) and singly coordinated without hydrogen ion ((FeO)PO₃) as reported by Tejedor and Anderson (1990) [183]. Daou et al., (2007) found that at the 3 pH, dihydrogen phosphate ions (H₂PO₄⁻) form a singly-protonated bi-nuclear phosphate compound on the magnetite nanoparticles surface. Thus, the interaction of phosphate with hydroxyl sites and specifically with Fe³⁺ ions situated on the surface. This interaction facilitates the swift development of a surface layer, impeding the dissolution of magnetite nanoparticles into the surrounding water [184]. Chowdhury et al., (2010) examined the impact of phosphate concentration for removing arsenic from synthetic as well as groundwater. From the experimental outcome, it was found that the percentage value for removing arsenic decreases, if phosphate concentration at the fixed adsorbent concentration is increased. The variation of percent values for elimination of As from synthetic and also from ground water with the concentration of PO₄³⁻ ions is as illustrate in Fig. 5(a) and (b). Magnetite-maghemite nanoparticles exhibit the

capability to reduce arsenic levels by less than 50% of the initial amount from an artificial water sample with a PO₄³⁻ concentration exceeding 3 mg/L. Furthermore, in the case of natural groundwater containing arsenic levels of 1.13 mg/L and a phosphate concentration surpassing 5 mg/L, magnetite-maghemite nanoparticles can lead to an arsenic reduction of less than 60% from the initial concentration [171].

7.2 Effect of zinc

In the basic medium, the addition of Zn²⁺ to the magnetite nanoparticles enhances the arsenic removal efficiency. Although Zn²⁺ doesn't impact the elimination of arsenic, presence of magnetite in Zn²⁺ containing solution (with 8.0 pH) can significantly affect the arsenic removal. Yang et al., (2010) have found that for 3.3mg/L of Zn²⁺, As(III) concentration decreases much more quickly, and was found below the MCL value within 10 hours, as shown in Fig. 6. Also, arsenic removal by magnetite nanoparticles increases with the addition of Zn⁺ from 66% to more than 99% for As(V) and from 80% - 95% for As(III) from an original As amount of about 100µg/L at a pH value of 8.0. At neutral or alkaline pH, zinc- arsenic complex behaves as metal-ligand complexes with stronger adsorption capacity [185].

7.3 Effect of bicarbonate and silica

Like phosphate, bicarbonate in water has a role in the adsorption of arsenic onto magnetite nanoparticles. Although, if no competing ion (*e.g.*, bicarbonate) is present, magnetite nanoparticles are sufficient for arsenic removal below to the standard value advised by MCL, although this increase in the concentration of HCO₃⁻ reduces the adsorption capability. For a fixed concentration of magnetite, arsenic removal efficiency decreases with the concentration of bicarbonate, and time for the adsorption also increases. As an illustration, after adding 8.2mM of HCO₃⁻ into a sample,

Fig. 5 Impact of PO₄³⁻ ions on (a) percentage removal of As (b) percentage removal of As at a pH value of 6.5 from arsenic rich underground water (initial arsenic concentration: 1.13 mg/L). Reprinted with due permission from Elsevier copyright 2010 (Chowdhury et al., 2010) [171]

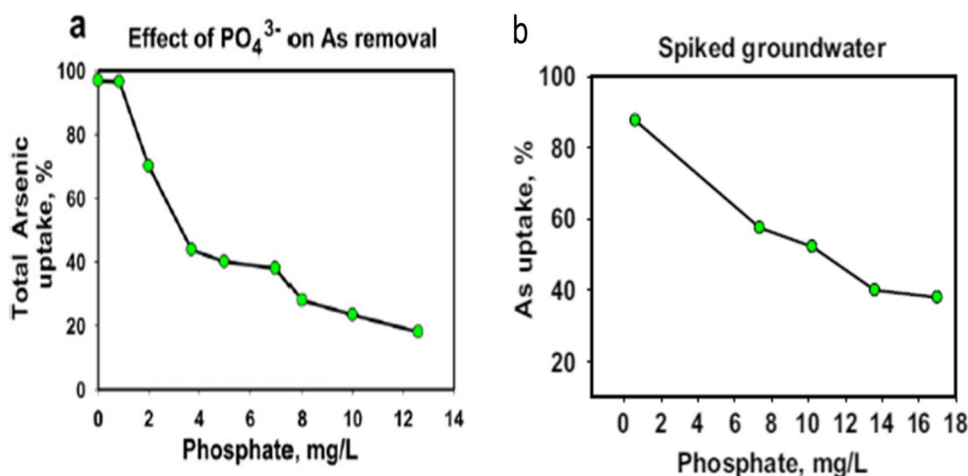
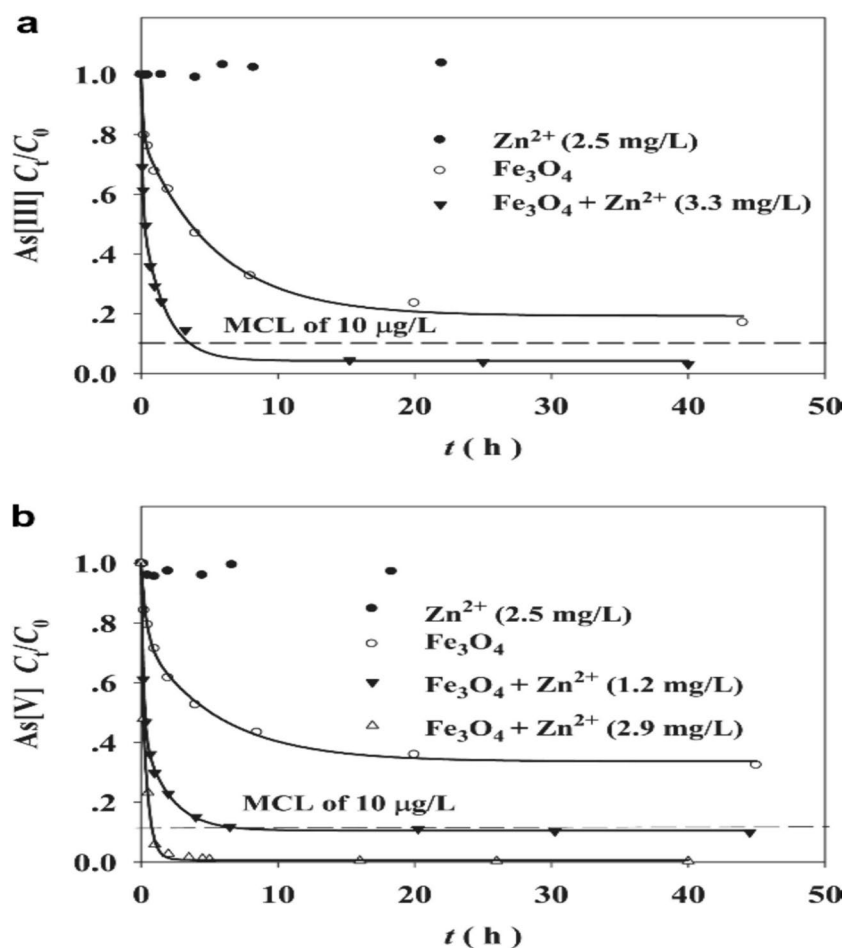


Fig. 6 Impact of Zn (II) ions⁺ on the elimination of (a) As(III) as well as (b) As(V), from nanoparticles of magnetite (Fe_3O_4) at a pH value of 8.0. Reprinted with due permission from Elsevier copyright 2010 [185]



it decreases the As(V) adsorption to 25.3 $\mu\text{g/L}$ as well as As(III) to 14.4 $\mu\text{g/L}$ from the agreed upon values of 48 $\mu\text{g/L}$ and 51.2 $\mu\text{g/L}$, respectively [186]. Similarly, in other studies 0.5g/L magnetite nanoparticles reduce the arsenic concentration to MCL in about half an hour. But when 100mg/L of HCO_3^- is added, the identical quantity (0.5g/L) of magnetite nanoparticles reduces arsenic concentration to MCL in about 1.5 hours for As(V) and 1.0 hour for As(III) [187]. The range of total carbonate in the ground water is about 0.5-8 mM, therefore carbonate can play a vital role in removing arsenic by occupying some of the sites on magnetite nanoparticles which were otherwise due for arsenic [187, 188].

7.4 Effect of ferrous/ferric ion

The efficacy for As elimination can be improved by introducing ferric chloride, which prompts the iron (III) hydroxides content in solution. This, in turn, causes expansion in available surface area for arsenic adsorption. As most natural water contains a fraction of few mg/L of di or trivalent iron, the addition of ferric chloride is a common practice to

enhance arsenic removal [189]. According to this study, the existence of 0.5mg/L ferric ion within the solution increases As(V) as well as As(III) adsorption by 12% related to 0.1g/L of magnetite nanoparticles in 1 hour. A similar but less effective behavior has been observed by ferrous ions. But post treatment removal from the water is a problem during the use of ferrous/ ferric ions and the iron hydroxides containing arsenic are hard to remove using low field magnets.

7.5 Effect of particles concentration

As the concentration of magnetite particles increases, both the total arsenic adsorption and adsorption rate increases. This could be accredited to formation of surface sites. The affinity of magnetite nanoparticles has been found similar for both As(V) and As(III) [187]. Shipley et al., (2009) have shown that concentration was reached below 10 $\mu\text{g/L}$ for As(V) as well as As(III) within 1 hour and 30 minutes, respectively by utilizing 0.5g/L of magnetite nanoparticles in water [190].

7.6 Effect of pH

In general, As(III) adsorption is pH independent while low pH values are more favorable for As(V) adsorption. Which is caused by difference in their charges. With a further increase in pH (up to approximately 8), more OH⁻ groups are generated on the surface of magnetite. As(III) predominantly acts neutral till pH 8. Consequently, its adsorption remains unaffected by changes in pH, as detected by Yean et al., (2005) [32]. They explored the influence of pH on As adsorption using magnetite with size 20 nm and 300 nm. They determined that the rise in pH doesn't influence As(III) adsorption onto both 20 nm and 300 nm nanoparticles. Additionally, when pH raised the maximum As(V) adsorption capabilities for 20 and 300 nm magnetite dropped.

Dixit and Hering (2003) explored sorption of As(III) onto magnetite exhibited an increase at lower pH values (< 9), while this sorption decreased at highly alkaline pH levels. A considerable sorption of both As on magnetite was obtained via Parga et al., (2005) when eliminating As from water in Mexico using electrocoagulation [191, 192].

The adsorption decreases as the pH increases and a small variation in As(V) adsorption was detected in within pH range of 2–6.5 [171, 193], which decreased sharply

above pH 7. Zhang et al., (2010), and Turk et al., (2010) showed similar pH dependency on adsorption efficiency by magnetite nanoparticles. They reported that adsorption of the arsenic on the nanoparticles increased with increasing pH for geothermal water and a highest adsorption was observed at natural pH value [170, 180]. Chowdhury et al., (2010) explored the pH influence on As eradication through a combination of magnetite-maghemite nanoparticles. They found that the uptake of both As(III) and (V) decreased with increment of pH. The efficiency of As(V) uptake dropped to below 10% when the pH surpassed 10 [171]. Additionally, the percentage of As(III) elimination experienced a rapid decline when the solution's pH reached 9 or higher, as illustrated in Fig. 7.

Khodabakhshi et al., (2011) similarly highlighted the impact of pH variation on arsenic elimination via magnetite nanoparticles, noting that the shift in pH from 3-9 distinctly impacts the process. The elimination of As(III) was discovered to be 59%, 77%, 82%, and 39% at pH of 3, 5, 7 and 9, respectively [167]. Chunming and Puls (2008) also confirmed the pH impact on As(V) adsorption by using eight commercial available magnetite for arsenic removal. The maximum degradation was noted below pH 5.6 – 6.8 [194].

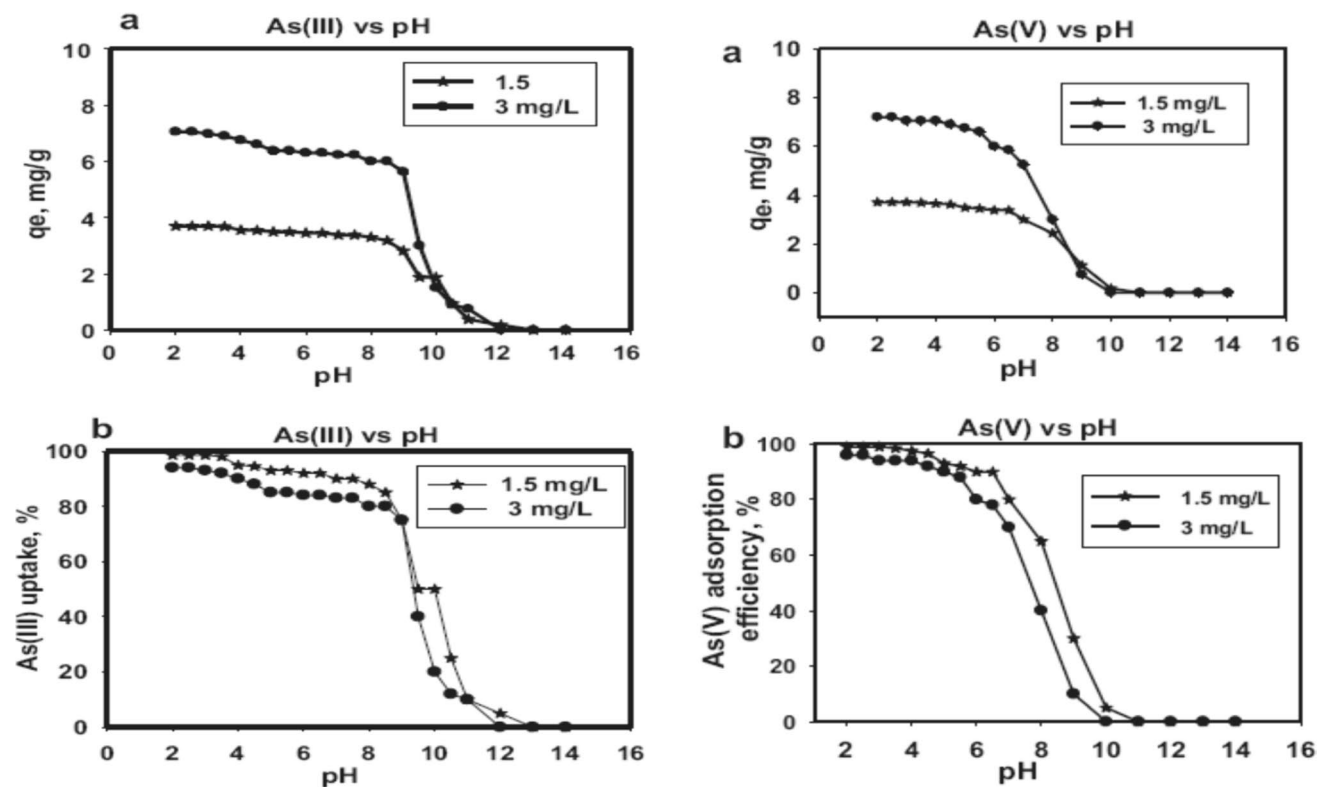


Fig. 7 Influence of pH on adsorption. Reprinted from permission from [171]

7.7 Impact of ionic strength

The ionic strength serves as an important factor affecting the adsorption efficiency of magnetite. Shipley et al., (2009) undertook adsorption experiments employing potassium nitrate (KNO_3) solutions of 0.01 M, 0.05 M, and 0.1 M, all at pH 8.0. At 0.1 M, the adsorption of both arsenate and arsenite measured $42.80 \mu\text{g/L}$ and $45.60 \mu\text{g/L}$, correspondingly, after 1 hour. The adsorption of arsenate and arsenite exhibited a 4% reduction upon going from 0.01 M to 0.1 M. This decline in arsenic adsorption with rise in ionic strength may be due to two possible reasons: (1) A rise in ionic strength could end up resulting in a reduced electrostatic double layer covering the magnetite, promoting aggregation, and diminishing the accessible surface area for adsorption. Given that the double layer can contract due to increased ionic strength and considering the negatively charged nature of the arsenate ion at a pH of 8, repulsion between arsenate and the surface of magnetite nanoparticles might occur; (2) Presence of competing species like potassium and nitrate [190].

7.8 Effect of temperature

Arsenic removal increases with rising temperature. This phenomenon is due to the solubility of arsenic species or alterations in surface characteristics, which, in turn, heightens the kinetic activity within the solution. This increased kinetic activity leads to a higher rate of contact between arsenic and the magnetite nanoparticles. However, beyond a certain optimal temperature, a reduction in arsenic removal efficiency is observed. This decline is accredited to the excessive rise in kinetic energy, causing dissolution of adsorbed arsenic. Shipley et al., (2009) investigated impact of temperature variations going from 20°C to 30°C on arsenic adsorption. They discovered that the adsorption of both type of As was notably greater at 25°C and 30°C compared to 20°C . Specifically, the quantities of adsorbed arsenate were $37.20 \mu\text{g/L}$ and $46.90 \mu\text{g/L}$, while for arsenite, the values were $42.60 \mu\text{g/L}$ and $49.30 \mu\text{g/L}$, for temperatures of 20°C and 30°C after 1 hour.

Moreover, there was minimal statistical disparity between degradation of As(V) and As(III) [190]. Comparable temperature-related tendencies in arsenic removal were shown by Pokhrel et al., (2008) and Chandra et al., (2010) [152, 195].

7.9 Impact of initial arsenic concentration

Primary concentration also affects the elimination efficiency of As by magnetite nanoparticles. Khodabakhshi et al., (2011) showed a investigate to comprehend the

influence of initial arsenic concentrations on its removal efficacy. The results revealed that under pH 7, and a nanoparticle dose of 5 g/L , the removal percentage of As stood at maximum of 79%, for 10 mg As/L and 16%, for 200 mg As/L initial concentration. This underscores the inverse relationship between arsenic removal and its initial concentrations. This is due to finite adsorption sites on a fixed quantity of nanoparticles. Consequently, as adsorbent content increases, the percentage removal of the adsorbate decreases due to the fixed capacity of available adsorption sites [167].

Using magnetite nanoparticles, Chowdhary and Yanful (2011) investigated how the initial As concentration affects the adsorption capacity of both Arsenic. Their results illustrates that when the starting concentrations of when both Arsenic inclined, the effectiveness of arsenic removal gradually decreased. When employing a constant adsorbent— 20 nm magnetite—at a dose of 0.4 g/L , this pattern was seen [193].

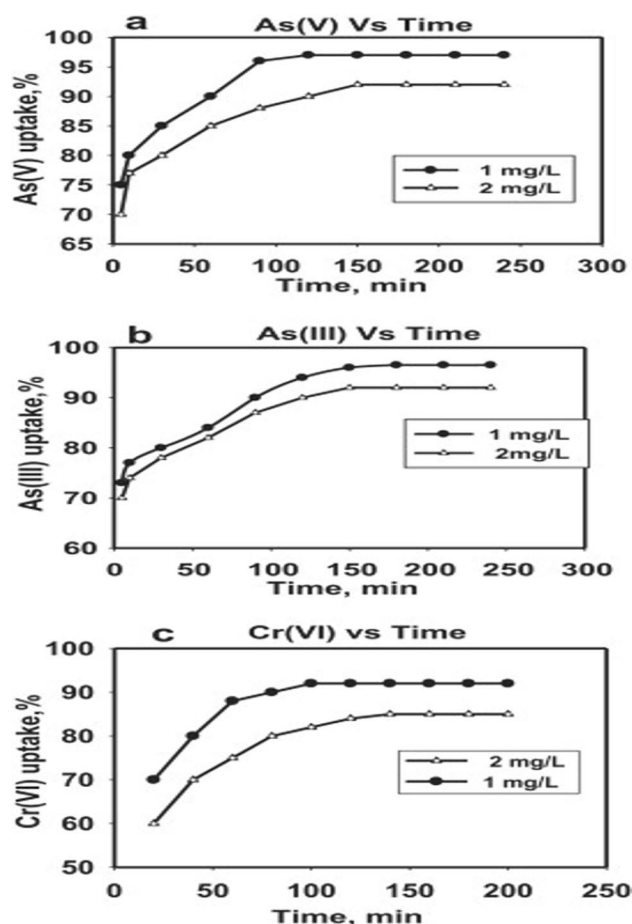


Fig. 8 (a) Impact of time for As (V) (b) Impact of time for of As (III); (c) Impact of time on uptake (%) of Cr(VI) . Reprinted from [171]

7.10 Effect of contact/agitation time

An essential factor in the effectiveness of arsenic removal is contact time. Many investigators have reported different contact times using different magnetite nano-systems. Chowdhury et al., (2010) conducted a kinetics investigation into the adsorption of As(V) and (III). They accomplished this by manipulating the duration of contact within magnetite-maghemite and solutions, between 10 to 240 minutes. The study utilized an adsorbent concentration of 0.4 g/L, with metal content set at 1 and 2 mg/L. The outcomes of this analysis, specifically elimination of both Arsenic at varying amount, are visually represented in Fig. 8(a, b). The results indicated a rapid uptake of the metals within the initial 10 minutes. The swift adsorption facilitated by the magnetite-maghemite combination is accredited to external surface interactions, making it convenient for arsenic to reach and engage with active adsorption sites. At pH of 6.5, equilibrium for various initial concentrations of both As types was attained in approximately 3 hours. At this equilibrium state, the efficiency of ejection for As(V) stood at 98% as well as for As(III) 92% was observed [171].

Türk et al., (2010) have found 60 minutes as optimum time for arsenic removal. Their findings revealed a significant reduction in As, dropping from 300 µg/L to 12.2 µg/L in just 15 minutes [170].

7.11 Effect of magnetite nanoparticles size

Surface area increases as the particle size goes to nanometer range. The fraction of atoms exposed at the surface increases giving rise to an excess energy, consequently, nanoparticles are thermodynamically metastable compare to macro crystalline materials. Therefore, nanoparticles with a higher surface energy are more prone to adsorb the molecule onto their surface to decrease the free energy [164, 196]. Mayo et al., demonstrated a decline in magnetite size, from 300 to 12 nm, resulted in a substantial enhancement in adsorption for As(III) as well as As(V), reaching 200 times the initial capacity [11].

Moreover, desorption of As from magnetite nanoparticles also depends upon their size, and a hysteretic behavior has been observed in desorption studies. According to the observation, desorption hysteresis was more severe in smaller particles, which is explained by the binding of adsorbed arsenic. As a result of this process, magnetite nanoparticle surfaces are uniformly covered in highly stable iron-arsenic complexes [11, 164].

8 Challenges and future recommendations

Adsorption is an efficient way to eliminate As from materials, however there are some issues that need to be resolved, such agglomeration in magnetite nanoparticles because of

their high magnetism [197]. The usage of suitable composites can help to lessen this. To evaluate the toxicity and possible health effects of these materials on people and the environment, extensive study is also necessary. Subsequent efforts must focus on expanding arsenic removal methods to encompass additional harmful contaminants in wastewater while maintaining vital minerals. Magnetite iron oxide nanomaterials need to be further improved in terms of their adsorption capacity and rate. This can be accomplished by adjusting the synthesis conditions to optimize active adsorption sites, optimizing the shapes and sizes of the particles and pores, or adding the appropriate functional groups to the surface based on the As characteristics. Therefore, more investigation is required to clarify the ambiguities surrounding the efficacy and hydrodynamic stability of magnetite nanoparticles in intricate real-world water matrices under controlled circumstances (e.g., low initial As content, neutral pH). In order to properly apply iron-based adsorption techniques in natural environmental settings, leaching studies are also required.

It is crucial to investigate magnetite utilization and its nanocomposites in removing of different heavy metals and contaminants from wastewater. The world's chemists, engineers, and biologists should work in unison to expedite efforts to commercialize these water purifying methods. These materials' proven capacity to be reused offers hope for affordable water treatment technologies.

9 Conclusions

This review indicates that magnetite could serve as a potential approach for addressing As-contaminated pure water. Magnetite nanoparticles present large adsorption capacity for arsenic, and are low-cost material. The primary mechanism underlying the remediation of arsenic-from water through magnetite appears to involve the adsorption on the hydroxide sites located on the magnetite surface. Therefore, magnetite nanoparticles can also be reused for further water purification because the arsenic adsorption is highly pH dependent. These nanoparticles being magnetic, are ideal sorbent as compared to their bulk counterparts or other material.

Numerous factors influence the adsorption of As by magnetite nanoparticles. These include the concentration of the adsorbent, the existence of typical water ions, and various factors like pH, ionic strength, and temperature. Arsenic absorption is noted to escalate with higher concentrations of the adsorbent and prolonged exposure time. The findings indicate that magnetite nanoparticles exhibit comparable adsorption tendencies for both As(V) and As(III), showcasing their tendency to attract both forms of arsenic. The existence of ions in solution, beside arsenic, can affect arsenic

adsorption. The addition of ions like silica and bicarbonate decreases arsenic adsorption, while the ferric and ferrous ions increase adsorption. The efficiency also rises with the contact time and dispersion of magnetite. The acidic and almost neutral pH are the most suitable condition for the (V) and As(III). As for the temperature, the ejection/ % first increases with temperature upto 30°C and then decreases. For the increase of arsenic content, the arsenic removal decreases. Magnetite nanoparticles can be separated by either filtration or magnetic separation due to their well-defined crystallinity. Also, the magnetite nanoparticles in different complex form show strong removal efficiency for arsenic, and can be parted by low magnetic field from the solution. Using nanomaterials for arsenic removal, magnetic separation systems utilizing magnetite nanoparticles may be at the frontier area of research. Nanoscale magnetite and their magnetic separation may provide long term environmental benefits.

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