# Chapter 21
# Nanoparticulate Iron Oxide Minerals for Arsenic Removal from Contaminated Water

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**Abstract** Groundwater contamination with arsenic (As) is a global environmental and human health problem affecting over 200 million people worldwide, with low to high concentrations of As via drinking well water. Therefore, remediation of As-contaminated water has been under discussion over the last 3 to 4 decades given its highly toxic and carcinogenic properties of As compounds, particularly inorganic arsenite and arsenate species. Several types of sorption techniques have been used to remove As from water such as clay minerals, biochars, metal oxides (e.g., iron oxide minerals), microbes and algae. This chapter provides: (1) insights on the significance of nanoparticulate iron (Fe) oxide minerals (such as nano-ferrihydrite, nano-goethite, nano-magnetite) for their efficiency in the removal of As from contaminated water; (2) develops critical understanding for several As removal methods, compares their potential for As remediation, and critically examines the properties and effectiveness of nanoparticulate Fe oxide minerals to remove As in drinking water or wastewater; and (3) implication of the nanotechnology in remediation of As-rich water. This chapter also elucidated the mechanism of As removal using Fe-oxide nanoparticles in detail.

**Keywords** Groundwater · Health · Nanoparticles · Remediation · Water treatment

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## 21.1 Introduction

Globally, arsenic (As) is recognized as one of the most toxic geogenic pollutant, which is released into groundwater and makes its pathway to humans through As-contaminated drinking water and food. Arsenic related health risks have been reported in more than 100 countries throughout the world, especially in the developing countries including Pakistan, Bangladesh and India (Hussain et al. 2021; Natasha et al. 2021). Natural processes including the weathering of parent material, geothermal waters and vulcanization and anthropogenic activities such as mining and smelting, metallurgy, fossil fuel burning, use of As-containing pesticides, herbicides, and crop desiccants have led to pollution of soil, groundwater, surface water with varying concentrations of As (Abbas et al. 2018; Bundschuh et al. 2022; Hussain et al. 2020a; Shaheen et al. 2022).

Chronic As exposure, particularly through drinking water, has been related to a number of detrimental health effects, including arsenocosis, cardiovascular and haematological effects, cancers of skin, bladder, kidneys and neurological problems (Aftabtalab et al. 2022; Shahid et al. 2017). Environmental and regulatory authorities have taken a serious stance on As in water due to its devastating effect on human health. The World Health Organization defined a maximum allowable limit for As in drinking water of 10 $\mu g\ L^{-1}$ (WHO 2011) and recently in the Netherlands a debate on setting a new limit of 1 $\mu g\ L^{-1}$ has been started for As in drinking water (Ahmad et al. 2020).

Several treatment technologies (e.g., chemical precipitation/flocculation, adsorption, ion exchange, reverse osmosis, and electrodialysis) have been evolved for removing As from contaminated water (Khan 2020; Natasha et al. 2021; Shaheen et al. 2022; Shakoor et al. 2018; Singh et al. 2022; Yadav et al. 2017). Membrane techniques such as nanofiltration and reverse osmosis, for remediating As in surface water and groundwater have been used over the past few decades (Amen et al. 2020; Basu et al. 2014b; Pan et al. 2021; Shaheen et al. 2022; Shakoor et al. 2016b). These techniques have some advantages such as high removal efficiency, convenience of use and reduced detrimental sludge buildup throughout the operation. The initial capital and running expenditures are high like extremely high pressure in membrane filtration to drive tainted water through the membrane requires energy and cost. Furthermore, in membrane processes concentrate discharge, membrane fouling, and flux reduction are unavoidable. Similarly, electrodialysis can eliminate As from water and other impurities, but it leads to a large amount of insoluble coagulant on the cathode (Sawood and Gupta 2018).

Because of its low cost, high efficiency and convenience of use, nanoadsorption may be one of the most promising technologies for eliminating As and other hazardous metal (liods) from contaminated water (Nazri and Sapawe 2020; Niazi and Burton 2016b; Pal et al. 2018; Patel et al. 2022b; Shaheen et al. 2022; Singh et al. 2022). Iron-based sorbents have been investigated and found to be effective in removing As from water (Ali et al. 2018; Shaheen et al. 2022). Granular ferric hydroxide (GFH) and nano zero-valent Fe are the two adsorbents that have been commercially produced on a significant scale with high As removal efficiency. However, most of these adsorbents are rarely used in practical field applications due to the existence of interfering ions in water despite the fact that they have high efficiency for removing As in water (Luo et al. 2018). Zero-valent iron is the most studied nanomaterial for water filtration. It is one of the most popular nanosorbents because it captures the broadest range of environmental pollutants such as halogenated organics, pesticides, arsenic, nitrates, and heavy metals (Table 21.1). Common anions such as chlorides, nitrates, and sulfates have little effect on As adsorption by the Fe-based sorbents because of the unique chemical interaction between As and Fe. Phosphate has been demonstrated to effectively compete with As, especially with arsenate (As(V)) for adsorption sites, thus decreasing adsorption capability of Fe oxides (Singh et al. 2015).

Similarly Rashid et al. (2020) reported that 99.57% of the As was removed with nZVI by adsorbing As species on the surface of Fe nanoparticles. Furthermore, Wu et al. (2019) found that As(V) adsorption was found to fit well with pseudo-first and pseudo-second order kinetic models, suggesting that removal of As(V)

**Table 21.1** Nanoparticulate iron oxide minerals, iron oxide minerals, and modified Fe oxide nanoparticles potential to remove arsenic (As) from As-contaminated water

| (Nano) iron oxide minerals | pH | Sorbent dose ($g\ L^{-1}$) | Arsenic (M) | Adsorption capacity (mg/g) | | References |
|---|---|---|---|---|---|---|
| | | | | As(III) | As(V) | |
| Ferrihydrite | 4.6, 9.2 | 2–40 | $2 \times 10^{-4}$–0.028 | 266 | 111 | (Baragaño et al. 2020; Gallegos-Garcia et al. 2012; Shaheen et al. 2022; Xu et al. 2022) |
| Ferrihydrite | 7 | 0.44 | 0–$2 \times 10^{-3}$ | – | 87 | |
| Goethite | 7 | 0.44 | 0–$5 \times 10^{-4}$ | – | 442 | |
| Goethite | 2–10 | 4 | $6.6 \times 10^{-6}$ | – | 0.50 | |
| Hematite | 2–10 | 4 | $6.6 \times 10^{-6}$ | – | 0.50 | |
| Goethite | 1.5–2.5 | 2.5 | $3 \times 10^{-5}$ to $1 \times 10^{-2}$ | – | 15 | |
| Goethite | 7.5 | 5 | $10^{-6}$, $10^{-3}$ | 0.374 | 0.449 | |
| Magnetite | 6.5 | 5 | $10^{-6}$, $10^{-3}$ | 0.206 | 0.253 | |
| Hematite | 7.3 | 5 | $10^{-6}$, $10^{-3}$ | 0.265 | 0.827 | |
| Granular ferric hydroxide (GFH) | 6.5–7.5 | – | 0.1 mg/L | – | 1.1 | (Sorlini et al. 2015) |
| β-FeOOH nanoparticles | 7.5 | – | 20 mg/L | – | 120 | (Sun et al. 2013) |
| Magnetite | 5.0 | – | 70 mg/L | 16.56 | 46.06 | (Feng et al. 2012) |
| Magnetite-maghemite ($Fe_3O_4$–γ-$Fe_2O_3$) | 2.0 | – | 1.5 mg/L | 3.69 | 3.71 | (Chowdhury et al. 2011) |
| Nano-Fe/oyster shell | 6.8 | – | 1.8 mg/L | 0.9 | – | (Fan et al. 2015) |
| $FeCl_3$ treated chestnut shell | 9.0 | – | 100 mg/L | 0.9 | – | (Targan and Tirtom 2015) |
| $Fe_3O_4$ coated wheat straw | 6–8 | – | 28 mg/L | 3.9 | 8.1 | (Sharma and Bhattacharya 2017) |

with Fe nanoparticles synthesized from eucalyptus involves a physical and chemical process adsorption. By promoting almost complete immobilization of As(V) species, Fe nanoparticles do not convert As(V) to the more toxic As(III) unless the Fe-material is exposed to a low pH and $E_h$ (Xu et al. 2022). After the diffusional surface adsorption and internal diffusional adsorption reach equilibrium, Fe(III) in the Fe nanoparticles is coordinated with As(V) to form monodentate chelating ligands and bidentate binuclear complexes (Table 21.1). Subsequently, co-precipitates form and accumulate into corrosion products on the surface of iron nanoparticles (Baragaño et al. 2020; Xu et al. 2022). So, keeping in view the importance and implications of Fe-oxide nanoparticles this chapter briefly reviews various As removal methods, compares their potential for As remediation, and critically examines the properties and effectiveness of nanoparticulate Fe oxide minerals for removing As from water.

## 21.2 Technologies for Arsenic Removal from Water

Arsenic removal technologies must meet a number of basic technical requirements, including robustness, environmental-sustainability, the ability to provide water supply systems all year, taking current climate change scenarios into account, and the required physicochemical and microbiological quality (Rahman et al. 2014). Main aim is to develop a conceptual framework for As removal that takes into account the presence of different As species such as As(III) and As(V) in the aquifers. If As removal is more critical and complex due to its hydrogeochemical behavior in water bodies, utilization of a membrane filtration procedure (reverse osmosis or nanofiltration) without an As(III) oxidation step (traditional or alternative approaches) can be employed (Seyfferth et al. 2010; Shakoor et al. 2016a).

## 21.3 Traditional Techniques

Physical exclusion is a method of eliminating dissolved As and other particulate components by passing them over synthetic membranes that are permeable to some, but not all dissolved substances. These membranes may remove dissolved As from the flowing water but this is an expensive method that involves high material and synthesis cost with high operation costs.

Coagulation filtration and lime softening are both inexpensive, but ineffective methods (~90%). Aluminum carbonate adsorption is one of the most efficient and cost-effective technologies (>95%) (Hoque et al. 2017). Under various experimental settings, many hybrid inorganic–organic adsorbents containing thiol groups were produced by altering activated alumina (AA) with mercaptopropyl-functionalized silica. The insertion of thiol groups improves the adsorption capability of the hybrid adsorbent for As(III), while maintaining the advantages of AA for As(V) adsorption (Postma et al. 2017).

### *21.3.1 Physicochemical Technologies for Arsenic Removal*

Although the above-mentioned traditional As removal techniques are well documented, some of them have recently gained popularity. New technologies for eliminating As are now being researched extensively. These methods concentrate on low-cost methods for improving the efficiency and cost-effectiveness of As removal in small water systems. Adsorption, for example, has been improved by introducing novel chemical oxidation processes and enhancing or employing new adsorption materials. The majority of these methods rely on As(III) oxidation followed by filtration through porous media to remove As via adsorption and coprecipitation (Mondal and Garg 2017).

### *21.3.2 Biological Methods for Arsenic Removal*

Many of the above chemical processes can be catalyzed by introducing bacteria to enhance bio-scavenging activity, but nothing is known about As bio-scavenging from water yet. Depending on the physicochemical state of the environment, some As compounds are extremely soluble. Arsenic species determines their toxicity and bioavailability, which is determined by microbial alterations such as reduction, oxidation, and methylation. Commercially viable and environmentally-friendly ways for removing As in water are chemical or adsorption based approaches. Understanding the metabolism of As in bacteria can aid in the prevention of As contamination in water bodies (Basu et al. 2014a; Hussain et al. 2021).

The majority of research, however, has been carried out at the pilot-scale and must be expanded and examined at large scale to determine their viability and potential for remediating As-contaminated waters. Various As remediation processes (such as oxidation/reduction, precipitation/dissolution, and adsorption/desorption) and biological mechanisms (oxidation, reduction, methylation, and thiolation of As species) (Hussain et al. 2019, 2020b). In certain reactions microbes play an important role in remediating As-contaminated water but the limitation of these process must be known that microbes can regulate the change in the $E_h$ and pH of the aquifers thus interfering the As hydrogeochemical cycling (Crognale et al. 2017).

## 21.4 Production of Nanoparticles and Their Implications

Nanoparticles are advanced materials in nanotechnology, and can be defined as the physical and chemical modification of substances to produce materials with specific features and properties that can be used for a variety of applications with a size of less than 100 nm (Badetti et al. 2021). Nanoparticles have features or functions that are distinct from bulk materials, such as thermal, electrical, chemical, optical, medicinal, and agricultural engineering, information, and communication (Khan 2020; Mushtaq et al. 2020). Traditional methods for synthesizing NPs, such as pyrolysis and abrasion, have a number of drawbacks, including surface creation flaws, limited productivity, high cost, and high energy demand (Table 21.2). The majority of the nanoparticles created are commonly used in photocatalytic dye removal techniques (de Souza Trigueiro et al. 2021). Toxic chemicals are frequently used in chemical syntheses such as sol–gel and chemical reduction processes, which result in harmful by-products and contamination of precursors.

As a result, it was discovered that developing ways to synthesis NPs results in nanoparticles that are clean, non-toxic, and ecologically benign (Siddiqui et al. 2019b). Green syntheses have been extensively documented in numerous publications for their adsorption capacity and effectiveness for pollutants removal such as As, Cr, and other PTEs when compared to other traditional approaches (Table 21.2). These green synthesized NPs do not include any dangerous chemicals and are made using non-toxic technologies to generate clean and environmentally friendly NPs on a wide scale (Sreeja et al. 2015). Biosynthesis components such as enzymes and microorganisms can act as capping or reducing agents, lowering the cost of the synthesis process and eliminating the need for significant energy consumption (energy saving). As previously stated, a great range of biological resources can be produced by synthesizing nanoparticles from microbes and plant extracts, such as Several authors have recently discovered methods based on microbial synthesis that mediate the biological creation of nanoparticles advantageous for the removal of contaminants, pharmaceuticals, product manufacturing, and other applications in their research and plants (Yan et al. 2015b; Yang et al. 2018).

Plant extracts can rapidly decrease metal ions, reducing the time required to synthesis NPs when compared to other biological sources. Depending on the plant variety and concentration of phytochemicals utilized, NPs might be created in minutes or hours. This benefit is clear when employing plants, as the synthesis of different nanoparticles takes time, but other natural sources create NPs quickly (de França Bettencourt et al. 2020). The disadvantage of microbes is a major issue while creating nanoparticles that require sterile settings. The costs of handling microorganisms, such as skilled personnel and cost scaling, are typically prohibitive (Dildar et al. 2022).

## 21.5 Technology for Nanoparticles Biosynthesis

Because of their numerous chemical and physical features, nanoparticles made by biological or known biosynthetic processes are becoming a popular synthesis approach. The necessity to generate environmentally-friendly nanoparticles in materials synthesis has drawn the attention of researchers all over the world to the integration of nano and biological technologies. The capabilities of this technique have been extensively researched, particularly in the synthesis of inorganic compounds (Table 21.2). Metal nanoparticles mediated by microorganisms and plants are the subject of

**Table 21.2** Iron NP synthesis techniques and their product morphology, advantages, and disadvantages

| Techniques | | Advantages | Nanoparticles morphological description | Disadvantages | References |
|---|---|---|---|---|---|
| Physical (8% of total nanoparticles production) | Electron beam lithography | Well-controlled interparticle spacing | Spheres and irregular spheres | Problematic in controlling the size of particle | (Soenen et al. 2009) |
| | Deposition of gas phase | Easy to execute | Spheres and rods | Requires expensive and highly complex machines | (Cuenya 2010) |
| Biological (2% of total nanoparticles production) | Microbial incubation | Small platelets, spherical or rod-like spheres, irregular spheres | Small platelets, spherical or rod-like spheres, irregular spheres | Slow and laborious | (Narayanan and Sakthivel 2010) |
| Chemical (90% of total nanoparticles production) | Oxidation | Narrow size distribution and uniform size | Irregular elongated and small spheres | Ferrite colloids of small size | (Lin and Samia 2006) |
| | Hydrothermal | Particle size and shapes are easily controllable | Elongated, compact rregular spheres, and numerous shapes | High pressure and reaction temperature | (Wu et al. 2008b) |
| | Sol–gel method | Aspect ratio, precisely controlled in size, and internal structure | Spheres, irregular spheres, porous and nonporous spheres, or spindles | High permeability, weak bonding, low wear resistance | (Laurent et al. 2008) |
| | Chemical co precipitation | Simple and effective | Spheres | Inappropriate for the synthesis of high untainted, precise stoichiometric phase | (Wu et al. 2011) |

ongoing research. The NPs are less harmful and environmentally-beneficial. Green chemistry can refer to a wide range of microorganisms, including bacteria, fungus, and plants (Niazi and Burton 2016a; Saunders et al. 2018; Yan et al. 2015a).

Nanoparticle creation requires three primary components that should be tailored to the green chemistry area, namely H solvents appropriate as synthesis media, a moderate reducing agent environment, and no harmful compounds are created. Synthetic methods have been shown to have major negative consequences for organic solvents, not only for the environment but also for individuals. Nanoparticles are safe for many manufacturing uses, thus research and development necessitate cross-sectoral usage of environmentally friendly and biocompatible processes to generate them (Singh et al. 2022). Microorganisms and plant extracts can thus produce nanoparticles that can be used as eco-friendly nano factories as building blocks and biomolecules.

## 21.6 Biocompatible Green Reagents Synthesis Biopolymers

Non-toxic synthetic biocompatible materials have been studied for the creation and stability of magnetic nanoparticle-polymer composites. Zhang et al. (2020) reported that water-soluble starch to stabilize bimetallic Fe/Pd nanoparticles in this experiment. Starch is a hydrophilic polymer made up of around 20% amylose that was discovered to be helpful in dispersing and stabilising iron nanoparticles in this study. Another work used a redox-based hydrothermal technique to make magnetite ($Fe_3O_4$) nanoparticles from the biopolymer sodium alginate utilising $FeCl_3.6H_2O$ and urea as starting ingredients. The sodium alginate nanoparticles had a homogeneous spherical shape with an average diameter of 27.2 nm. Ahmad and Mirza (2018) first created well-dispersed magnetite ($Fe_3O_4$) agar nanocomposites by co-precipitating Fe(III) and Fe(II) ions.

Further Patel et al. (2022a) reported on the creation of Fe nanoparticles utilizing ascorbic acid (vitamin C). Aqueous ascorbic acid (vitamin C), which lowers transition metal salts to their corresponding nanostructures, was used to make core–shell iron and copper nanoparticles. Similarly, Savasari et al. (2015) employed ascorbic acid to make stable zero-valent iron nanoparticles that self-assemble into chains, with individual particles measuring 20 to 75 nm in diameter. Furthermore, ascorbic acid has been employed as a nanoparticle functionalizer and stabiliser. In one work, superparamagnetic iron oxide nanoparticles were coated with ascorbic acid (vitamin C) and subsequently functionalized to generate stable dispersions for medical purposes. The coated nanoparticles revealed spherical particles with an average particle size of 5 nm in transmission electron microscopy (TEM) pictures (Sreeja et al. 2015).

***Amino acids***: Wet-chemical co-precipitation was used to prepare amine-functionalized magnetite nanoparticles, according to Krishna et al. (2012). Functionalization with L-lysine amino acids produced a highly crystalline magnetite phase (in situ). Similarly, the effect of pH on zero-valent iron production was studied using various amino acids such as L-glutamic acid, L-glutamine, L-arginine, and

L-cysteine. Hemoglobin and myoglobin are two different types of hemoglobin. Single-phase chemical reduction yields stable iron nanoparticles at room temperature, according to a study. The synthesized particles' size distribution ranged from 2 to 5 nm, and they were found to be crystalline. This method of making bioconjugate nanoparticles for biological applications could be a useful and important technical approach.

## 21.7 Arsenic Removal Using Nanoparticulate Iron Oxides

Nanoparticluate Fe oxides mineral-based sorbents have received significant attention because of their high As removal efficiency, eco-friendly nature, and ease of synthesis and availability (Table 21.2). The research was previously focused on the synthesis of new nano-Fe adsorbents with high As adsorption potential (Xu et al. 2022). Nanoadsorbents produced from (nano) Fe-oxide minerals, nano zero-valent Fe, Fe-based bimetallic oxides, and Fe-impregnated composite adsorbents has been explored for their application for As removal under different conditions (Aftabtalab et al. 2022; Baragaño et al. 2020; Rashid et al. 2020; Siddiqui et al. 2019b).

Environmental remediation employs certain metal nanoparticles (NPs) with adsorptive capacities such as $TiO_2$, ZnO, and Ag NPs (Zhu et al. 2019b). However, due to their potent photocatalytic properties, these metal nanoparticles are better suited for use in ceramics, optics, chemistry, biology, electronics, and other domains. Many other compounds have been reported as above to have a great affinity for As, but Fe oxyhydroxide has gotten the most attention because of its ease of usage (Saravanan et al. 2021). Ferrous and ferric salts, which are generated by hydrolysis and oxidation processes, can precipitate as akaganeite ($\gamma$-FeOOH), goethite ($\alpha$-FeOOH), ferrihydrite ($Fe_{10}O_{14}(OH)_2$), and patina as shown in Fig. 21.1 (Cantu et al. 2016).

## 21.8 Arsenic Removal Adsorption Process

### 21.8.1 Coagulation/Flocculation

Colloidal solid particles in As-contaminated water initially coagulate because the ions appear as hydrolyzed species in the Stern layer of the colloidal particles ($Al_3$ or $Fe_3$ ions). Electrolytic coagulation has the similar mechanism of As removal as the addition of single coagulant (Mohamed Noor et al. 2021). Dissolved As ions react with hydrolyzed species in the stern layer to produce Fe-As(V) ($FeAsO_4$) or aluminum-As(V) ($AlAsO_4$), which is adsorbed on the coagulum. This phenomenon is referred to as precipitation or co-precipitation (Siddiqui et al. 2019b).

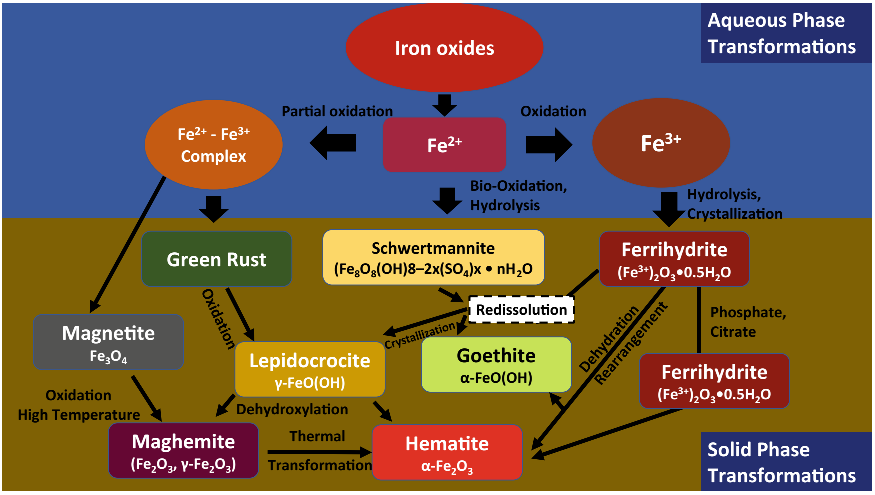

**Fig. 21.1** Different iron oxide minerals and their transformations pathways in aqueous and solid phase

### 21.8.2 Ion Exchange Method

Further advancement to coagulation method methods includes the exchange of ion between different species. Thus the addition of Fe-oxides and Al(OH) flakes, for example, exhibit a strong attraction for dissolved As. Arsenic is drawn to adsorption sites on the solid surface and remediated from the solution phase. Ion exchange is a one-of-a-kind form of adsorption, and it is frequently regarded as such. Ion exchange is the reversible displacement of adsorbed ions by dissolved As species on a solid surface. Other sorts of adsorptions result in stronger, reliable compounds that has more half-life with larger surface area that can be easily removed by using other coagulants. In the adsorption process, the adsorbent is the most significant factor. Adsorbents with a porous structure and a high surface-to-volume ratio are effective (Basu et al. 2014a; Singh et al. 2015).

To remove As from water, various adsorbents (natural and synthetic sources) have been developed, such as polymeric resins, activated carbon, ion exchange resins, and hydrous metal oxides, such as activated alumina, metal-supported coral limestone, hematite ore, and porous resins loaded with crystalline hydrated zirconia. The Fe oxide has a stronger ability to absorb As from water than activated alumina in fixed bed systems. Adsorption is commonly thought of as a method of removing As (Aftabtalab et al. 2022; Saravanan et al. 2021; Shaheen et al. 2022). It is influenced by pH, sorbent pretreatment, and the presence of other ions (sulfate, chlorides, etc.). Organic components in aqueous solutions can reduce As removal considerably. This could be explained by competing effects of coexisting solutes on adsorption, such as surface complexation processes, inner and outer layer complexes along with string affinity for As sorption sites. The phosphate ion is a frequent competitor in the As adsorption process due to chemical characteristics similar to As(V) (Mandal et al. 2018; Niazi et al. 2017).

## 21.9 Adsorption of Arsenic on Nano-Iron Enabled Minerals

### *21.9.1 Nano Iron Oxide Minerals for Arsenic Adsorption*

Iron oxide minerals such as goethite, hematite, limonite, ferrihydrite, and magnetite has been reported to be used as adsorbent for As removal (Table 21.1). Goethite ($\alpha$-FeOOH) is a Fe oxide mineral formed of two FeO(OH) octahedral double bands sharing edges and corners to form a 2:1 octahedral tunnel partially connected by H-bonding (62.9%), Fe and $O_2$ (27%), 10.1% of O and $H_2O$ (Fig. 21.2). This sample contains acicular crystals with grooves and edges. Hematite ($Fe_2O_3$) is a mineral composed of 70% iron and 30% oxygen. The cations are octahedrally coordinated, and the structure is based on hexagonal oxygen atom closest packing (Nazri and Sapawe 2020). The most common and important iron ore is hematite, which can be found in rocks.

The ability of Fe oxide minerals to absorb As has been studied in a number of ways. Goethite is the best iron oxide mineral for adsorbing As in water. The adsorption of As(V) on goethite in water was studied by Mamindy-Pajany et al. (2009) as a function of pH and ionic strength. Goethite retains a maximum amount of As(V) in acidic pH conditions with adsorption of As(V) on the goethite surface is unaffected by ionic strength. Moreno-Jiménez et al. (2012) reported that similar results for As(III) and As(V) adsorption on goethite in water. Synthetic goethite has a capacity of 5 mg $g^{-1}$ of As adsorption at pH 5.0 (Chowdhury et al. 2016). To assess the As adsorption capacity, Siddiqui and Chaudhry (2017) reported that in a batch experiments with synthetic goethite at pH 1.5–2.5 and various ionic strengths (0.02–0.15 $mol^{-1}$ NaCl). A Langmuir isotherm was used to fit As adsorption to goethite. Ionic strength and pH have modest effects on adsorption capacity at lower pH values. Sulfate ions obstruct As removal from water by competing with As(V) for adsorption sites on the goethite surface. Hematite's potential to remove As(V) from aqueous settings has also been investigated. To fit the adsorption of As(V) on hematite, Langmuir isotherms were used (Siddiqui et al. 2019b; Singh et al. 2015).

Adsorption of As(V) is preferred electrostatically above hematite PZC (point of zero charge) (pH 7.1). Adsorption fell below pH 4.2 due to hematite breakdown and reduction in the number of adsorption sites between 3 and 11 on the pH. The adsorption and desorption behaviour of As(V) and As(III) on doublet ferrihydrite was studied by Liu et al. (2020) that showed As(V) and As(III) significantly adsorbed on Fe-oxide materials. The As(V) is thought to have a stronger affinity for Fe-oxide surfaces than As (III) (Xu et al. 2022).

Magnetite has been utilized as adsorbents to remove As from water in series of batch sorption experiment. However, the kinetics of As(V) adsorption on goethite

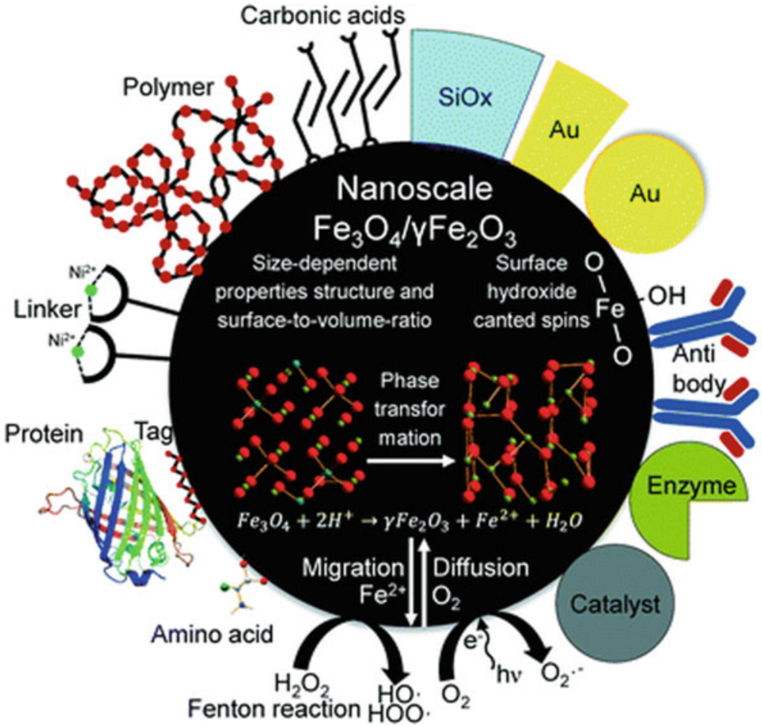

**Fig. 21.2** Sorption pathways for the iron oxide nanoparticulate minerals (Reproduced with permission from Publisher from Schwaminger et al. (2017). This article is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported Licence and open access to use material)

revealed two states (Yadav et al. 2017); (i) the first high rate of adsorption could be due to As(V) adsorption at more accessible spots on the goethite's outer surface, whereas (ii) the second low rate of adsorption could be due to As(V) slowly diffusing into the pores of the goethite particles due to goethite (Balint et al. 2020).

### 21.9.2 Arsenic Adsorption on Nanoparticulate Iron Oxide Minerals and Effect of Various Factors

The speciation of As in solution is influenced by the pH of the solution. The pH-dependent distribution of As(V) and its hydrolyzed species. While As(V) species are only stable in the right pH range, for example, pH 2 for $H_3AsO_4$, pH 2–7 for $H_2AsO_4$, pH 7–11 for $HAsO_2$, pH > 12 for $AsO_3^{4-}$, At pH 9, As(III) is stable as $H_3AsO_3$, pH 9–12 in $H_2AsO^{3-}$, pH 12–13 in $HAsO_2^{3-}$, and pH > 13 in $AsO_3^{3-}$ (Xua and Lia 2020). Each species of As is known to have a particular affinity for the

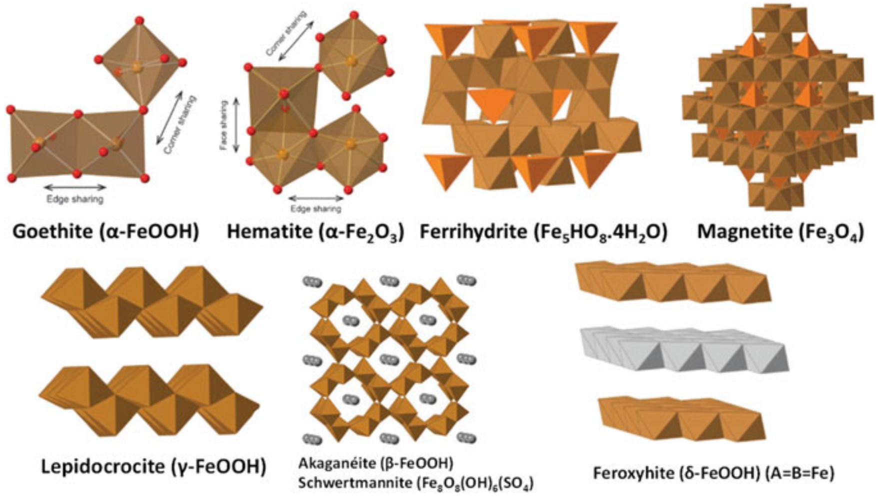

**Fig. 21.3** Structural demonstration of the different nanoparticulate iron oxide minerals (modified from Scheinost and Singh 2022)

surface of Fe-oxide minerals. As a result, the chemisorption capacity of As on Fe-oxide mineral surfaces changes depending on the As species and hence the pH of the solution altered (Fig. 21.3). The adsorption of As(V) by Fe-oxide minerals declines with increasing solution pH and peaks at very low pH values, whereas As(III) adsorbs at these pH values and peaks around pH 8.5. (Koomson and Asiam 2020). The pH of the solution has an impact on the surface charge on Fe-oxide mineral particles. The surface is negatively charged in the pH range above PZC (Khan 2020; Pan et al. 2021).

Because the charge of the adsorbent and the adsorbate have the same sign in the pH range above PZC, there is an electrostatic repulsion between the As species and the surface of the Fe-oxide material, that results in low sorption for As species (Adebayo et al. 2020). The amount of As adsorption onto Fe oxide minerals is reduced when competing anions are present. In the presence of phosphate ions, for example, As(V) and As(III) adsorption on goethite is greatly reduced, while some sites are far more selective for As(III) than for phosphate. Other anionic components, such as sulphates, chlorides, or natural organics, can impair the efficacy of As removal by adsorption utilizing Fe-oxide rocks as the adsorbent (Doherty et al. 2021). The conflicting effects of coexisting solutes on As adsorption, such as surface complexation processes, can explain this. Because of its molecular closeness to As(V) and its abundance in natural water, phosphate is a common competitor in the As adsorption process (Kobya et al. 2020).

### *21.9.3 Adsorption of Arsenic by Iron Oxide Minerals in Water*

Several Fe-oxide minerals have been investigated for their ability to adsorb As from contaminated water. A large percentage of As was removed. The elimination of arsenic is greatest at acidic pH and minimal at alkaline pH (Kamei-Ishikawa et al. 2017). At pH 3–6 and an As concentration of 13.35 $\text{mmol}^{-1}$, the greatest removal of As(V) using hematite as an adsorbent was almost 100% (Koomson and Asiam 2020). Rahim and Haris (2015) reported that the ability of natural hematite to remove As from drinking water using batch and column experiments. It has been discovered that as hematite grain size decreases, removal efficiency increases. Nitrate ions had little influence on As(V) uptake, however phosphate ions significantly slowed it down. Natural hematite, can be used as a sorbent to extract As from water, but it is more effective than hematite (Guo et al. 2014). The physical and chemical features of mineral powders, such as particle size, specific surface area, surface active sites, and microscopic surface morphology, are known to influence As adsorption on Fe oxide minerals.

## 21.10 Arsenic Adsorption Mechanisms on Nanoparticulate Iron Oxide Minerals

The extended X-ray absorption fine structure (EXAFS) spectroscopy was used to investigate As(V) adsorption on Fe-oxide minerals (Herath et al. 2020; Palansooriya et al. 2020). On the Fe-oxide surface, the As(V) complex in the form of the bidentate binuclear inner sphere was shown to be the thermodynamically most favorable and consequently most numerous species. However, there are some inconsistencies in the production of bidentate mononuclear and monodentate complexes, leaving As(V) adsorption on Fe-oxides unsolved. On dried samples, Fourier transform infrared (FTIR) spectroscopy was used to analyze development of bidentate binuclear complexes (Zama et al. 2018; Zhu et al. 2019a). The hydroxyl groups on the Fe-oxide surface generate the As complex, according to the findings. The hypothesized mechanism consists of two steps: (i) the creation of a monodentate surface inner sphere complex with a high adsorption rate, (ii) followed by slow ligand exchange and the formation of a bidentate inner sphere complex (Cui et al. 2018; Wu et al. 2018).

Covalent bonds between adsorbed ions and reactive surface functional groups are defined as inner sphere complexes (Pintor et al. 2020). In a 1:1 stoichiometric ratio, the complexes of the inner spherical surface can form monodentate complexes (e.g., -Fe-$OAsO_3H$) or bidentate complexes in a 1:2 stoichiometric ratio (Sobh et al. 2019). Most As(V) and As(III) oxanions replace the two separately coordinated -OH groups on the surface of Fe-oxide minerals, forming the Fe-O-AsO(OH)-O-Fe and Fe-O-As(OH)-O-Fe dinuclear bridging complexes (Dixit et al. 2016). Arsenite prefers two-coordinate surface OH groups, whereas As(V) prefers three-coordinate surface OH groups. The $HAsO_2^{2-}$ ion takes part in the ligand exchange reaction, displacing mono-coordinate surface hydroxyl groups and adsorbing as dinuclear species to iron oxide minerals (Zhu et al. 2020). The predominant binding mechanism for As(V) adsorption to goethite is still the bidentate binuclear complex. The age of the surface-covering oxides influences the three forms of As(V)-goethite surface complexes. The ligand exchange reaction of $H_2AsO_2^{4-}$ with surface OH groups create monodentate complexes with exceptionally low surface occupancy (Li et al. 2017; Zhao et al. 2021). After the second ligand exchange event, the production of bidentate surface complexes dominates the adsorption of As(V) at large surface loadings.

On goethite and ferrihydrite, As(V) forms intraspheric bidentate complexes. Monodentate complexes can form on crystalline goethite under these conditions, but only at very low surface coverage. The proportion of monodentate bonds diminishes as As(V) coverage of amorphous iron oxide crystals increases, and bidentate binuclear bridge complexes become the dominant adsorbed complexes. pH and As species have a big impact on how As interacts with iron oxide surfaces. On ferrihydrite, the largest As(V) adsorption occurred between pH 3.5–5.5, while the maximum As(III) adsorption occurred between pH 8 and 10. Variable charge characteristics and As species on the surface of Fe oxide minerals are blamed for these tendencies (Almeida et al. 2020). Electrostatic attraction and surface complexation between As species in solution and Fe(II) and/or Fe(III) hydroxides on minerals are thought to be involved, resulting to As adsorption on iron oxides via PZCs (Saravanan et al. 2021; Siddiqui et al. 2019a; Wu et al. 2011, 2008a; Xu et al. 2022).

The As adsorption of Fe oxide minerals has been studied using a variety of techniques including FTIR spectroscopy, X-ray absorption spectroscopy (XAS), and scanning electron microscopy coupled with energy dispersive spectroscopy (SEM–EDX). As a direct method for examining As adsorption processes on Fe oxide minerals, ATR-FTIR spectroscopy has been proposed. Anions displace $OH^-$ and/or $H_2O$ off the surface, resulting in differences in band intensity after and before adsorption, which can be detected by FTIR spectroscopy (Yu et al. 2018).

## 21.11 Conclusions and Future Perspectives

This chapter reviews some important As removal technologies with a particular emphasis on nanoparticulate Fe oxide minerals for As remediation in water because conventional or membrane-based techniques are complex, expensive, and cause secondary pollution in form of sludge production. As a result, selecting the most appropriate treatment plan is critical in order to meet increasingly stringent quality standards of various impurities and toxic ions, such as As in water. The decreasing number of water sources suitable for public supply, the increasingly stringent nature of drinking water quality standards, and the potential impacts of climate change on the quantities of potable water highlight the significance of alternative, sustainable, and low-cost As removal technologies.

Until now, the majority of previous research has been directed into developing new adsorbent materials with high adsorption capabilities. Furthermore, nanomaterials have been investigated for As adsorption in recent years although nanoparticles could agglomerate in water, reducing adsorption and removal efficiency. As a result, loading nanoparticles onto appropriate supporting bio- or geo-materials is becoming a viable technique, with the benefits of high reactivity and ease of water separation. Adsorbent-based technologies must be developed further to be utilized in the field in a sustainable manner. More research is needed to discover the local and frequently available resources to produce Fe nanoparticles to accomplish the sustainability of nanomaterial synthesis. Understanding the molecular mechanisms involved in Fe oxides nanoparticles efficiency to remove As is essential for determining the environmental fate of adsorbed As and its safe disposal. Further studies in the future should focus on green synthesis techniques to mediate Fe nanoparticles production at large scale. It is important to improve Fe oxides nanoparticles stability. Some previous studies have reported that the biosynthesized Fe nanoparticles are less hazardous than designed nanoparticles. Furthermore, a full risk evaluation of Fe oxides nanoparticles, green fabricated Fe NPs should be conducted considering their toxicity and fate, transport, dissolution, and kinetics in the environment. The green nanotechnology techniques discussed in this chapter could provide a powerful tool and solid foundation for the manufacturing of a wide range of biological or functionalized Fe oxide nanoparticles that can be used in the development of novel products for use in environmental remediation and restoration programs.

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