



Review

A Review of Theoretical Knowledge and Practical Applications of Iron-Based Adsorbents for Removing Arsenic from Water

Thi Hai Nguyen ¹, Tien Vinh Nguyen ^{2,*}, Saravanamuthu Vigneswaran ^{2,3,*} , Nguyen Thi Hoang Ha ⁴ 
and Harsha Ratnaweera ³

¹ VNU Key Laboratory of Geo-Environment and Climate Change Response, University of Science, Vietnam National University, Hanoi 120080, Vietnam

² Faculty of Engineering and IT, University of Technology Sydney (UTS), Ultimo, NSW 2007, Australia

³ Faculty of Sciences & Technology (RealTek), Norwegian University of Life Sciences, P.O. Box 5003, NO-1432 Ås, Norway; harsha.ratnaweera@nmbu.no

⁴ Vietnam Japan University, Vietnam National University, Hanoi 101000, Vietnam

* Correspondence: tien.nguyen@uts.edu.au (T.V.N.); s.vigneswaran@uts.edu.au (S.V.)

Abstract: Groundwater contamination by arsenic (As) is a serious issue in many countries, particularly in middle- and low-income nations. High concentrations of As in drinking water and groundwater cause serious health problems. Numerous technologies, such as ion exchange, oxidation, coagulation–flocculation, phytoremediation, membrane filtration, and adsorption, have been applied to remove As from the water environment. Among these technologies, adsorption is the preferred method in low- and middle-income countries due to its affordability, high efficiency, and ease of design, operation, and maintenance. Along with this technology, a variety of materials have been developed and investigated for their ability to remove As from water environments, though iron-based adsorbents have been reported to be the most effective. Thus, the aim of this paper is to briefly review the sources of As and the prevalence of As species in water bodies, as well as the currently available technologies for As removal. A detailed analysis of recent studies on iron-based adsorbents used to remove As from aqueous solutions in both laboratory conditions and field conditions is presented.

Keywords: arsenic; adsorption technology; iron-based adsorbent; theoretical knowledge; practical application; water environment



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1. Introduction

More than 30% of the world's population uses groundwater as a primary source of drinking water [1]. Groundwater sources in many countries, including India, Bangladesh, China, Cambodia, Nepal, Vietnam, Argentina, Bolivia, Chile, Mexico, and Brazil, have been contaminated with arsenic (As) [2,3]. Although the presence of As in bodies of water in various environments is mostly a result of naturally occurring processes (the weathering of minerals, soils, rocks, and sediments, volcanic processes, biological activities, etc.), the main reason for the remarkable increase in As concentrations in groundwater in today's world is human activity [1,4]. Pollution in groundwater affects more than 230 million people worldwide [1]. Most affected people (78% of exposure reports) are in the world's underdeveloped nations, which are mainly located in South Asia. All of these severely affected countries are classified as low- and middle-income nations, e.g., India, Bangladesh, Nepal, Vietnam, and Cambodia [2]. As-contaminated groundwater and its impacts on human health represent a major barrier to achieving the basic levels of the UN's sustainable development goals SDG-3 (i.e., ensure good health and promote well-being) and SDG-6 (i.e., clean water and sanitation for all). Target SDG 3.9 involves reducing the number of deaths and illnesses derived from hazardous chemicals in water, whereas Target 6.1 specifies achieving equitable access for all to safe and affordable drinking water. In low-

and middle-income countries, hundreds of millions of people depend on groundwater sources. Thus, As removal from contaminated ground water will be mandatory if these countries want to achieve the SDG-3 and SDG-6 targets prior to the specified deadline of 2030.

In order to achieve the above targets, As removal from the water environment is an urgent requirement in the contaminated areas. Recently, various techniques have been developed and applied to remove As, and these include ion exchange [5], oxidation [6], coagulation–flocculation [4], phytoremediation, membrane filtration [7], and adsorption [8]. However, the development of these technologies and their application to groundwater systems in low- and middle-income countries remains a significant challenge for researchers, manufacturers, and policymakers. Among the existing technologies, adsorption is the most suitable method for low- and middle-income countries because of its low cost, high efficiency, and the simplicity of its design, operation, and maintenance. With this in mind, a wide range of materials were used to develop adsorption filters, and their adsorption capacity for removing As from various bodies of water is the focus of this paper.

These materials are produced from different sources, including natural and synthetic materials as well as reused waste and/or by-products from agricultural and industrial processes [9]. Of note are the iron-based adsorbents, which are considered to be the most effective for As uptake from aqueous solutions [10,11]. The high affinity of iron for As ions makes it especially suited to remove this pollutant. Moreover, some of these iron-based adsorbents have been practically applied in filtration systems in order to provide a safer water source for human consumption [12–17]. Hence, through this review, the sources of As and the prevalence of As species in water bodies are documented. A review of current adsorption techniques using iron-based adsorbents for removing As from aqueous solutions is the main focus of this paper. An appraisal of current studies on the abilities of iron-based adsorbents to remove As from water, both in laboratory conditions and in practical applications, is presented in detail. This paper also discusses the advantages and limitations associated with these techniques, as well as the challenges involved.

2. Sources of Arsenic

Naturally occurring processes and human activity are the main causes of the presence of As in groundwater. The three primary natural processes are as follows: (i) the weathering of minerals, rocks, sediments, and soil; (ii) volcanic activity; and (iii) biological activity. The factors controlling these processes include the reducing and oxidizing conditions, the hydrogeological and morphological characteristics of the aquifer, and the abundance of degradable organic material. In particular, the leaching of As from As-bearing iron minerals (arsenopyrite (FeAsS), pyrite ($\text{Fe}(\text{SAs})_2$), realgar (AsS), orpiment (As_2S_3), and lollingite (FeAs_2 , FeAs_3 , and FeAs_5)) is considered to be a major cause of the presence of As in groundwater in many nations, including Bangladesh, Taiwan, India, Vietnam, Hungary, and Romania [2,18]. In South America, As contamination principally results from the dissolving of As from volcanic rocks, volcanic ash, and geothermal fluids into water sources and soils. Consequently, As contamination was identified at more than 100 aquifers across river basins in this region. For the Mekong and Red River Deltas in Vietnam, the main causes of naturally occurring As in groundwater have been traced to a number of hydrological factors, including the rapid sedimentation of alluvial materials, the geography of the delta, and the decomposition of sediments rich in organic matter, which leads to depleting oxygen levels at the bottom of alluvial sections of rivers [18]. In numerous parts of the United States, Thailand, Ghana, and other countries, mining and mining-related activities (Sn-mining, Au-mining, and Pb–Zn–Ag-mining) are also thought to be a factor contributing to As mobility in water [2,3]. This is because the creation of acid mine drainage leads to the dissolving of minerals and the releasing of As into the water. Moreover, leather-working, sheep dipping, the smelting of non-ferrous metals, the production of combustion products (wood, coal, and debris), pigments, leather preservatives, poisonous baits, catalysts, pyrotechnics, antifouling agents in paints, and

pharmaceutical substances, and the extensive use of pesticides, herbicides, and insecticides in industrial and agricultural practices also contribute to arsenic pollution and cause serious As contamination of the environment [19].

3. Arsenic Species and Their Toxicity to Human Health

As can exist in the environment in a number of oxidation states, including -3 , 0 , $+3$, and $+5$. Nevertheless, As is often found in natural water in the form of the inorganic compounds trivalent arsenite (As(III)) and pentavalent arsenate (As(V)) [2,20]. Due to its high solubility and mobility, As(III) is approximately 5–10 times more dangerous than As(V) [21,22]. Interestingly, the presence of different examples of As speciation in an aqueous environment is greatly influenced by pH and redox potential. For example, under slightly reducing conditions and at low pH ($pK_a < 9.2$), As(III) is found in the stable form H_3AsO_3 . The charged As(V) species ($H_2AsO_4^-$, $HAsO_4^{2-}$, and AsO_4^{3-}) predominate in oxidizing conditions within the pH range of 2.3–11.6, while H_3AsO_4 and AsO_4^{3-} are arsenate species that occur in very acidic and alkaline environments.

As derived from inorganic compounds is extremely poisonous and carcinogenic, and it can enter the human body through food and/or inhalation pathways [22]. The toxicity of As to humans depends on the chemical form and physical state of the molecules involved. In contrast to the inorganic As found in water, organic As compounds have a framework of carbon atoms that are less harmful to human health. Additionally, the effects of As on human health are contingent on the amount of As consumed as well as individual factors such as gender, age, health condition, personal sensitivity to As, dietary patterns, and lifestyle. As has an impact on the proper functioning of nearly all the body's organs, such as the gastrointestinal, cardiovascular, urinary, hepatic, dermal, hematopoietic, and neurological systems. The potential for developing cancer(s) in the human skin, lungs, liver, bladder, kidneys, and prostate has been shown to increase after long-term exposure to As [23–25].

4. Adsorption: A Promising Technology for Water Treatment

A number of advanced and conventional treatment techniques have been suggested for removing As from aqueous solutions, both in the laboratory and in real-life applications. The primary As treatment technologies include oxidation, coagulation–flocculation, phytoremediation, ion exchange, membrane filtration, and adsorption [6,26–30]. Each technique has its own set of advantages and disadvantages. For instance, the oxidation process has attracted interest because of its ease of use, low installation cost, high potential for treating large volumes of water, and effectiveness in removing As(III). However, the use of these oxidants might produce harmful and cancer-causing compounds, which is a drawback [6,29,31]. During ion exchange, the treatment process does not depend on the influent concentration and pH solution. Therefore, the pH solution does not need to be adjusted. However, the effectiveness with which this method removes As may be diminished by the presence of common anions such as SO_4^{2-} , F^- , and NO_3^- . Since As(III) is found as an uncharged ion in natural water, this procedure is not appropriate for As(III) anion removal. Furthermore, iron precipitates and suspended particles can rapidly clog the system and reduce the efficiency of the As removal [5,26,27,30].

Coagulation–flocculation, which is often used on a centralized water treatment scale, may remove 90% of As(V) from a solution. This method can be effective over a wide pH range. Its limitations include a costly initial capital investment, the requirement for an additional separation step, and the ineffectiveness of the process for eliminating As(III) ions [27,32]. Among these technologies, membrane filtration—a widespread solution—is used due to its outstanding characteristics, which include its high efficiency, its simplicity of operation, its applicability in both centralized and decentralized contexts, and the relatively small space it requires [29,33]. However, this technique is rather expensive (both in terms of capital and maintenance costs), uses a lot of electricity, and results in a lot of water

rejection. Another disadvantage is that the reverse osmosis membrane eliminates essential trace ions from the treated drinking water along with As and other contaminants.

Although phytoremediation technology is an environmentally friendly technique, its drawbacks include the large amount of space it requires and its reliance on the climate and plants [27,28,34]. Among the various As-removal methods, adsorption is widely recognized as the best alternative technique for decentralized treatment systems due to its cost-effectiveness and the simplicity of its design, operation, and maintenance [14,27]. Along with this technology, a variety of materials have been developed, and a large range of adsorbents have been investigated for their ability to remove As from water environments. Technology has also enabled the refinement of a wide range of adsorbents, all of which have been tested for their ability to remove As from water environments. Iron-based adsorbents have attracted the most attention for their exceptional performance in removing As. Although several iron-containing adsorbents have been reported to play a dual role in adsorption technology, serving as both oxidation agent and adsorbent, in practical applications, a pre-treatment step (an oxidation process to convert As(III) to As(V)) is necessary when a large amount of As(III) is observed in water.

5. Iron-Based Adsorbents: Excellent Adsorbents for Removing Arsenic from Water

Iron-based adsorbents have been extensively developed for the removal of As ions from aqueous environments due to their strong affinity with As. Tables 1–6 summarize the Langmuir maximum adsorption capacity (Q_{max}) of iron-based adsorbents towards As(III) and As(V). In the literature, popular iron-based adsorbents are grouped into the following six classes:

(i) Iron oxides/hydroxides: Among iron-based adsorbents, iron oxides/hydroxides are the most extensively investigated due to their high affinity to As and their accessibility [35]. The most well-known iron oxides/hydroxides are magnetite (Fe_3O_4), hematite (Fe_2O_3), maghemite ($\gamma-Fe_2O_3$), and FeOOH polymorphs (e.g., goethite ($\alpha-FeOOH$), akaganèite ($\beta-FeOOH$), lepidocrocite ($\gamma-FeOOH$), ferrihydrites ($Fe_{10}O_{14}(OH)_2$), and granular ferric hydroxide (GFH)). Furthermore, the polymorphs of the FeOOH group are normally synthesized from Fe(III) or Fe(II) salts by employing chemical precipitation methods. Banerjee et al. [36] report that a combination of granular ferric hydroxide (GFH) and akaganèite ($\beta-FeOOH$) serves as a commercial adsorbent on an industrial scale. Moreover, some natural iron-rich minerals, such as laterite, laterite soil, and siderite, are considered more promising and cost-effective materials for As adsorption [37–39].

Table 1. The Langmuir maximum adsorption capacities (Q_{max}) of iron oxides/hydroxides towards As.

Adsorbents	S_{BET} (m^2/g)	C_0 As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
Natural laterite (from India)	18.05	1–3	20	7.2	0.17	–	[38]
Laterite soil (from India)	15.36	–	20	7.2	–	0.18	[39]
Natural siderite (from Germany)	–	0.25–2		2.0	1.04	0.52	[37]
Natural laterite (from Vietnam)	155	0.1–25	7.5	7.0	0.51	0.58	[14]
Goethite (from the USA)	13	0.1–3	1.25	7.0	–	1.31	[40]
Goethite (from China)	9.02	–	1	7.0	–	1.18	[41]
Hematite (from India)	14.40	0–0.5	4.2	2.0	–	0.20	[42]
Hemantite (from Sinopharm Chemical Reagent Company, China)	4.16	–	1	7.0	–	0.85	[41]
Granular ferric hydroxide (GFH)	240–300	–		6.5	–	1.1	[36]
Nanocrystalline- Fe_3O_4	11.9	1–100	8	7.9	–	4	[43]
Mixed magnetite–maghemite nanoparticles	49	0.5–4	0.4	2.0	3.69	3.71	[44]

(ii) Iron-coated natural mineral adsorbents: The most popular natural mineral adsorbents include clay-group minerals (kaolinite, montmorillonite, bentonite, etc.), zeolite-group minerals (e.g., clinoptilolite, volcanic glass, and chabazite–phillipsite), sand, and

rock. Although these materials are very popular throughout the world, the negative charges on their surface structures means that they have only a low capacity to adsorb As. Consequently, numerous studies have attempted to load iron oxide/hydroxide onto such natural minerals and then investigate their ability to remove As [20,45].

Table 2. The Langmuir maximum adsorption capacity (Q_{max}) of iron-coated natural minerals towards As.

Adsorbents	S_{BET} (m ² /g)	Co As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
Iron–manganese-modified zeolite-rich tuffs	35.54	0.05–2	10	6.5	0.06	0.1	[46]
Fe-exchanged natural zeolite	–	0.1–20	20	–	0.05	0.1	[47]
Iron-coated sand	20.5	0.01–0.5	24	5.0	0.013	0.022	[48]
iron oxide-coated sand	10.6	0.1	–	7.6	0.041	0.043	[49]
Iron-modified light expanded clay aggregates	–	0.1–100	10	6.0	–	3.12	[50]
clay/Fe–Mn composite	55.45	10–400	2	–	–	120.7	[51]
Fe-modified clinoptilolite	–	0.1	0.5–12	6.6	–	0.0084	[52]
Polymeric Al/Fe-modified montmorillonite	172	10–100	3.33	9 for As(III); 4 for As(V)	19.11	21.23	[53]

(iii) Iron-coated bio-adsorbents: Iron-coated natural bio-adsorbents and iron-coated biochars are affordable, sustainable, and environmentally acceptable adsorbents with a remarkable ability to effectively remove As from water [54,55]. The most commonly used type of bio-adsorbent is agricultural waste sourced from rice husks, sawdust, corn straw, corn stems, maize cobs, orange peel, pomelo peel, etc. Biochar is a stable carbonaceous product that is typically produced by the pyrolysis process at temperatures between 300 °C and 1000 °C. Consequently, due to its abundance, inexpensiveness, and widespread availability, iron-loaded biochars or biosorbents are potentially effective adsorbents for removing As from water.

Table 3. The Langmuir maximum adsorption capacity (Q_{max}) of iron-coated bio-adsorbents towards As.

Adsorbents	S_{BET} (m ² /g)	Co As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
Iron-coated pomelo peel	5.43	0.05–20	1	7.0	11.77	15.28	[8]
Fe-modified corn straws	208	0.2–50	1	7.0	–	8.25	[56]
Fe ₃ O ₄ -coated wheat straw	4.73	1–28	0.5	6–8	3.90	4.02	[57]
Iron-loaded orange peel	–	0.01–50	10	7.0	–	37.5	[58]
Iron-coated honeycomb briquette cinder	–	0.10–0.50	0.26	7.5	–	0.96	[59]
Fe-modified corn straws	208	0.2–50	1	7.0	–	8.25	[56]
Fe(III)-coated rice husk.	–	1–75	4	4.0	–	2.5	[60]
Fe (III)-loaded chitosan hollow fibers	–	0.2–0.3	–	3.5	–	3.70	[61]
Iron oxide-amended rice husk char	77.3	0–2.5	1	6.8	–	0.6	[62]
Fe-coated cork granulates	–	1–40	2.5	–	4.9	4.3	[63]
Fe-modified biochar powder	123.8	1–30	2.0	7.2	13.5	–	[64]
HA/Fe–Mn oxide-loaded biochar	82.88	10–70	0.5	7.0	–	35.59	[65]
Magnetic biochar	139.1	20–500	2	3.8	–	24.9	[66]
Magnetic biochar	119.3	10–500	2	10.9	–	144.9	[67]

(iv) Iron-coated activated carbon: Activated carbon (AC) or granular activated carbon (GAC) has been employed as an adsorbent to remove various contaminants from aquatic environments due to its high specific surface area, high adsorption capacity, pore structure, and customizable surface functionalization [68]. Moreover, it has been described as the most widely used commercial adsorbent in both centralized and decentralized scenarios. However, the applications of GAC are limited by its high cost (which includes the cost

of its maintenance and replacement). In addition, the negatively charged surface of AC causes it to exhibit a low capacity for adsorption of As species. In order to improve its As adsorption capacity, several studies over the past few decades have attempted to load iron oxide/hydroxide onto activated carbons (ACs). Due to their strong affinity for As species, iron oxide/hydroxide particles are a significant component of these iron-based activated carbons and actively contribute to the removal of As. The main advantage of AC is its high surface area, as stated above. In order to combine their outstanding As adsorption properties, it was determined that mixing iron oxide/hydroxide and AC was a crucial strategy.

Table 4. The Langmuir maximum adsorption capacity (Q_{max}) of iron-coated activated carbon towards As.

Adsorbents	S_{BET} (m^2/g)	Co As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
GAC-Fe	876	0.1	0.1–0.8	6.0	–	1.430	[69]
Iron-impregnated GAC	–	0.5–10	2.8	7.0	–	1.95	[70]
Iron-modified AC	–	20–22	0.1–20	6.0	38.8	51.3	[71]
Fe(II)-loaded IAC	987	0.5–8.5	0.4	3.0	–	2.82	[72]
Fe(III)-loaded IAC	1231	0.5–8.5	0.4	3.0	–	4.67	[72]
Fe–Mn binary oxide-impregnated GAC	420.12	1–600	3.3	7.5	18.43	15.95	[73]
Fe–Mn–straw AC	507.5	0–350	1	3.0	75.82	–	[74]
AC modified with iron hydro(oxide) nanoparticles	632–1101	0.050	0.75	7.0	–	0.37–1.25	[75]

(v) Nanoscale zero-valent iron (nZVI) and nZVI-coated adsorbents: Nanoscale zero-valent iron (nZVI) has received a lot of interest for its efficiency as an adsorbent in the removal of numerous typical pollutants in aquatic environments [76]. The large surface area, extremely small particle size, and great in-situ reactivity of nZVI have made it a popular choice for removing As from contaminated water. nZVI can remove the majority of As through its co-precipitation with iron oxides/hydroxides that are created during nZVI corrosion (oxidation). Although nZVI has been celebrated as a promising material for As removal, its direct application in water treatment systems could cause material loss (due to its nanosize) and result in an enriched iron concentration in drinking water. In addition, the preparation procedure of nZVI is extremely complicated and time-consuming, and its cost is high. Therefore, in order to prepare nZVI on a large scale, it is essential to explore new methods for preparing nZVI at an affordable price while preserving its properties, such as its high adsorption capacity and high quality. Furthermore, pure nZVI is easily oxidized and difficult to preserve. Although borohydride is frequently used as a reducing agent in the preparation of nZVI, its cost is still high, and it may cause secondary pollution.

Table 5. The Langmuir maximum adsorption capacity (Q_{max}) of nanoscale zero-valent iron (nZVI) and nZVI-coated adsorbents towards As.

Adsorbents	S_{BET} (m^2/g)	Co As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
nZVI	–	2	0.1–1	7.0	12.2	14	[77]
nZVI-reduced graphite oxide-modified composites	100.65	1–15	0.4	7.0	35.83	29.04	[78]
sulfide-modified nanoscale zerovalent iron	37.8	10–100	0.5	7.0	89.29	79.37	[79]

Table 5. Cont.

Adsorbents	S_{BET} (m^2/g)	Co As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
ZVI-impregnated chitosan-carboxymethyl- β -cyclodextrin composite beads	12.1	1–20	3.33	6.0	18.51	13.51	[80]
nZVI stabilized with starch and carboxymethyl cellulose	–	0.25–10	0.3	7.0	12.2	14	[77]
Montmorillonite-supported nZVI	36.97	2–200	1.0	7.0	59.9	45.5	[81]

(vi) Iron-based layered double hydroxides (LDHs): LDHs have recently garnered increasing attention as adsorbents for removing As from water due to their uncomplicated preparation process, distinctive characteristics, and high affinity with As ions [82,83]. LDHs belong to the synthetic clay group and have numerous ionic layer structures in their galleries, including positively charged brucite-like layers and non-framework charge-compensating anions. As a result, the brucite layers of the material's positive charge can effectively react with As through the electrostatic attraction mechanism. The host anions (CO_3^{2-} and NO_3^- anions) in the interlayer regions exhibit a very high exchange with As anions in solution. Fe^{3+} ions are combined with one or two additional divalent cations, namely Mg, Mn, Ni, Zn, Cu, and Co, in the brucite-like layers to create most of the iron-based LDHs. The most popular co-precipitation method served to create iron-based LDH at various cation mass ratios.

Table 6. The Langmuir maximum adsorption capacity (Q_{max}) of iron-based layered double hydroxides (LDHs) towards As.

Adsorbents	S_{BET} (m^2/g)	Co As (mg/L)	m/V (g/L)	pH	Q_{max} (mg/g)		Ref.
					As(III)	As(V)	
Mn/Mg/Fe-LDH	72.5	4–270	1.0	7.0	56.1	32.2	[83]
Fe/Al-LDH	87.42	0.2–250	–	6.0	37.6	40.6	[84]
Mg/Fe-CLDH	145.3	50	1.0	7.0	–	50.24	[85]
ZnFe-LDH	11.9	2–100	0.2	7.0	–	96.91	[86]
Fe-Mg-LDH	273	30–750	5.0	6.0	–	194.5	[87]
Cu/Mg/Fe/La-LDH	134	1–15	0.2	6.0	–	43.5	[88]
Calcined Mg-Fe LDHs	287.4	206	1.0	6.5	–	202	[89]
Fe/Mn-C Layered Double Hydroxide Composite	170.51	5–100	1.4	–	45.43	37.84	[90]

6. Practical Application

Adsorbents for the removal of As from aquatic environments have recently been the subject of numerous studies; however, most of these were conducted in laboratories, and examples of practical applications in the real world are limited. The reason for this is that practical implementation is far more challenging than testing in a laboratory setting. The wide ranges of As concentrations in water, investment costs, maintenance costs, generation of As waste, disposal of exhausted adsorbents, socioeconomic conditions, awareness of local people, willingness of local communities to pay, and the perceptions of local people are all very real challenges that need to be addressed to successfully implement these adsorbents in practice. These factors determine the popularity of technology in general, and the implementation of adsorbents in particular.

In some practical projects, adsorbents are already being employed in filtration systems, and more studies are required to supply safe water sources in affected areas such as Bangladesh, India, and Vietnam. The overall objectives of applying materials in filtration systems are as follows: firstly, to eliminate As pollution from polluted water, and secondly, to provide clean, safe, and As-free water for people living in these impacted areas. Adsor-

bents are subjected to constant testing of their performance prior to being used in filtering systems at various scales, from household units large communities. For a household water filter, a capacity of at least 10 to 12 L/h is typical, as this is adequate for providing drinking water for a family of four to five members. It goes without saying that the types of water filtration systems required for household use are relatively modest, simple to use, and easy to maintain. The ability to function without an electrical power source is another great benefit of properly designed home filtration systems.

At the community level, filtration systems must be more intricately designed than at the household level as they require a larger space and must be powered by electricity. The number of individuals who benefit from community filtration systems can range from hundreds to thousands of people. Notably, the majority of filtration systems, regardless of scale, are developed utilizing one or a combination of diverse technologies, including oxidation, precipitation, coagulation, and adsorption. Some studies have applied adsorbents in community and/or household filtration systems to produce water free of As. Examples of these include activated laterite [13], natural laterite [14,15], iron-coated sand [91,92], iron oxide/hydroxide-coated chitosan [17], and zero-valent iron [93–95].

6.1. Practical Application of Iron-Based Adsorbents

India is renowned for having experienced significant As poisoning in recent decades. Because of this, scientists, politicians, policymakers, and water providers have worked intensively to identify suitable and effective adsorbents that can be applied in filter systems to remove As from contaminated water sources. A research group at the Indian Institute of Technology Kharagpur created the “low-cost laterite-based As filter” project for removing As from wells in West Bengal, India [13]. Treated laterite was prepared via acid and alkali treatment of raw laterite and then packed into the filter cartridges of both household and community filtration systems. In one study, the modified laterite adsorbent had a maximum adsorption capacity (32.5 mg/g) that was higher than those of expensive activated alumina, iron oxide-coated sand, and iron-based commercial adsorbents [13]. The filter cartridge was composed of many layers of materials, such as activated carbon, charcoal, sand, activated laterite, and raw laterite, which are utilized in both family and public contexts.

The daily capacities of the residential and community filtration systems were 40–120 L/day and 500–2000 L/day, respectively. This technique was successfully applied in 3 communities and 20 households in West Bengal that served more than 5000 people who were exposed to high levels of As contamination in groundwater (between 0.05 and 0.5 mg/L). After filtration, the As concentration in the output water greatly diminished and satisfied the WHO’s drinking water standards (As < 10 µg/L). As-, iron-, and coliform-free safe water was provided by the filter to the inhabitants of the affected area. The further benefits of this filtration system are as follows: (i) the adsorbent can be employed continuously over its five-year lifetime without the need for regeneration or backwashing; (ii) the exhausted adsorbent does not release any hazardous substances upon disposal; and (iii) the household filtration system can operate without electricity. As a result, it is still feasible to install these household filtration units in rural places or isolated villages that have either no electricity or a limited power supply.

A recent project carried out by a research team from the University of Technology Sydney together with their Vietnamese partners aimed to investigate the removal capacity of a low-cost adsorbent (natural laterite (NLTT) from Vietnam) as well as its As-removal performance in filtration systems applied to groundwater in affected regions of Vietnam [14,15]. The NLTT originated from the naturally iron-rich subsoil rock in a specific location in the Thach That area. After a simple production process, which included exploration, grinding, and washing, the NLTT was packed into water filtration systems. It is estimated that about 2000 people in the Red River Delta, Vietnam, are now supplied with safe water from the new household and community water filter systems (in childcare centers, health stations, cultural houses, temples, etc.) produced by this project. The results of a 7-month monitoring period revealed that after 6–7 months of operation, both the household and community

filtration systems could still successfully reduce the amount of As in the groundwater at the investigated sites to less than 0.01 mg/L, which is the limit for As in the Vietnamese and WHO drinking water guidelines. The results showed that all the water quality parameters (total Fe, Mn, Cu, Cr, Pb, Hg, Ni, Se, and Cd) for the effluent of the filtration systems also met the Vietnamese drinking water guidelines.

The community and household filtration systems were designed with capacities of 500 L/h and 10 L/h, respectively. The primary community filter included the following: (i) a combination tank for aeration and clarification processes in to remove both iron and As simultaneously; (ii) an up-flow adsorption column for removing As and other pollutants; (iii) an ultrafilter for the removal of microorganisms; and (iv) a storage tank. The adsorption column used in this project included sand (288 kg), NLTT (220 kg), and commercial GAC (140 kg). For the household filter, 15 kg of NLTT and 1 kg of sand was packed into the PVC pipe (140 mm in diameter and 650 mm in height). The already existing sand filters in each household were used to perform a pretreatment step to enhance the NLTT's capacity to remove As, Fe, and other pollutants. Most significantly, the household filter could operate without any electricity [15]. The exhausted adsorbents were replaced after 7 months' operation, following which they could be used as material for producing concrete for road construction.

Some synthetic adsorbents are also now being applied in practice. Confined metastable 2-line ferrihydrite (CM2LF), a synthetic nanocomposite adsorbent, was developed by applying the precipitation method to iron oxyhydroxide and chitosan [17]. Because it has a high iron content, an inherently porous structure, and a large surface area (172 m²/g), the maximum As adsorption capability of CM2LF is 100 mg/g. This material has been extensively used in a filtration system known as "Arsenic and Metal Removal by Indian Technology (AMRIT)" to purify the water in As-contaminated areas in India. The research team responsible for this project won the 2023 VinFuture Prize (under the "Innovators from Developing Countries" category) for their development of a low-cost filtration system to remove As and other heavy metals from groundwater. More than 600,000 people in Punjab, West Bengal, Uttar Pradesh, and other Indian states received As-free water thanks to this technology [17].

AMRIT now operates in a variety of contexts, including households, small communities, and large-scale water plants. During this project, four distinct designs for community filtration systems were created with relative capacities of 17,000 L/h, 8000 L/day, 12,000 L/day, and 1000 L/day. These community filters sufficiently delivered clean, As-free water to locals in affected areas with populations of 200, 250, 1000, and 5693. In two districts of West Bengal, over 160 units of a community filtration system were actively used in communes and even remote villages. In both West Bengal and Bihar, about 200 home filtration systems with 3 L/h capacities were installed [96].

Petrusevski et al. [92] evaluated the ability of iron-coated sand (IOCS) in both community (IHE ADART) and household filtration systems (UNESCO–IHE filter) to remove As from groundwater in southern Hungary and rural areas in Bangladesh, respectively. IOCS originated from groundwater treatment plants in the Netherlands, where the sand was naturally coated with Fe and Mn during its lifetime in these plants [16]. The Fe and Mn in IOCS play an important role in removing As because they have a high affinity with As. Because of the excellent performance of IOCS in laboratory experiments, it was packed into filter cartridges in water treatment plants. The community filtration system, with its capacity of 55 m³/day, consisted of a plate aeration tank and three pressure steel filters with an external diameter of 0.8 m and a height of 4 m. The second and third steel filters were filled with 2 m of IOCS, while the first steel filter was filled with 2 m of quartz sand. In the household filtration system, the IOCS was packed into a filter made of PVC pipe that was 66 mm in diameter and 500 mm in height. The monitoring results found that both the community and household filters could produce As-free water (As < 10 µg/L) for 18 months without the need to replace the adsorbent. Moreover, aeration and rapid sand filtration played important roles in the whole process and contributed to the high efficiency

in removing methane, ammonia iron, and manganese from tube wells in southern Hungary. The results documented by Petrushevski et al. [16] showed that the household filtration system was able to provide safe water for drinking and cooking for a typical family in rural Bangladesh. The household filter system was designed to operate under gravity, and therefore does not require a power supply. As a result, the operational and capital costs of this filtration system are low enough to be affordable for rural communities in Bangladesh. For treatment plants with a capacity of less than 8000 m³/day, the overall treatment costs are estimated to be less than EUR 0.10/m³ [91].

Recently, zero-valent iron (ZVI) has attracted interest due to its exceptional performance in eliminating As from water, and various studies have utilized ZVI to treat actual As-contaminated groundwater. The most commonly known and highly successful ZVI-based filter to date is the SONO filter. Around 30,000 SONO filters have been installed throughout Bangladesh, and they supply more than a billion liters of clean drinking water annually [94,97]. The SONO filter can supply As-free water for up to 8 years. The results indicate clearly that the filter can keep the As concentration in the filtrate below the standards required in Bangladesh. Another well-known study that involved field testing in Burkina Faso was carried out by Bretzler et al. [12]. The designed filtration systems were based on the SONO filter. It was observed that the filters required no maintenance during the 12-month trial period. Although these filters had been designed in the same way as the SONO filter, their removal capacity was not expected to be as high as that of the SONO filter.

The Kanchan filter (KAF), another commonly used ZVI-based filter, was developed and tested in Nepal. There are more than 5000 KAFs operating, providing services to between 35,000 and 50,000 individuals [95]. Despite the fact that their average As-removal capacity is 90%, the filters are rarely able to bring the As concentration in the filtrate below 50 µg/L. This is either because the contact time is too short or the influent As concentration is too high (>500 µg/L), both of which can result in unsatisfactory As removal. Another pilot study using ZVI at the community level was undertaken by the Indian Institute of Technology Bombay (IIT Bombay) [93]. A total of 58 filters were installed in locations throughout India, including Uttar Pradesh, Bihar, West Bengal, and Assam. The results confirmed that these filters could consistently produce safe water with an As concentration of around 10 µg/L. The estimated cost of producing 1 m³ of water (the average monthly requirement for a family of five) varies from INR 0.10 to INR 1.00 and includes the maintenance of the hand pump, iron replenishment, and filter-washing labor costs. It has therefore emerged as a cost-effective technology for India's rural areas.

Activated alumina, another common adsorbent, is currently being used in various filtration systems [98,99]. Through a partnership project between Lehigh University (United States) and Bengal Engineering and Science University (BESU, India), more than 200 community-level As-removal devices, each with a flow rate of 10 mL/min, have been deployed in India since 1997 [98]. In this filter system, 100 L of activated alumina is used in the filter cartridge. These systems provide daily As-free drinking water to almost 200,000 inhabitants in the affected area. The concentration of total dissolved solids, iron, and manganese in the treated water, as well as its hardness, also meet the drinking water regulations. Another project in India also deployed activated alumina in their As-removal units (ARUs). Kumar et al. [99] carried out a project involving 365 ARU filters installed in Uttar Pradesh, India. Each filter could provide As-free drinking water for approximately 200–300 households. These ARU filters incorporate the techniques of oxidation, co-precipitation, and adsorption. The results revealed that only 4 of the 200 tested ARU filters were able to reduce the As concentration to less than 50 µg/L. As a result, the project failed. Due to their socioeconomic circumstances, many of the families living in villages with ARUs were either denied access to the filtered water or chose not to use it. There were two main reasons for this: (i) the media (activated alumina) was unavailable in the local markets and (ii) the filters were easily clogged and required continual maintenance.

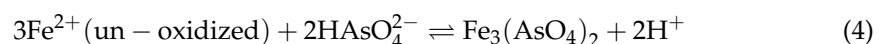
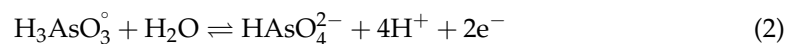
6.2. As-Removal Mechanisms of the Filtration Systems

The majority of treatment techniques used in both household and community filtration systems are based on the principles of oxidation, precipitation, and adsorption. These technologies are chosen because of the following factors:

6.2.1. Oxidation and Precipitation

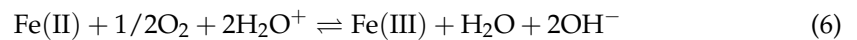
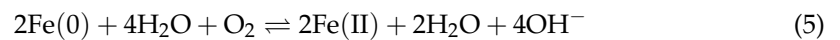
In groundwater, As is primarily present as As(III). According to Singh et al. (2015), it is found in the stable form H_3AsO_3 , which has a very low affinity towards adsorbent surfaces. Thus, it is crucial that filtration systems include oxidation processes. The oxidation process by which the neutral As(III) forms the negatively charged As(V) (H_2AsO_4^- , HAsO_4^{2-} , AsO_4^{3-}) increases the effectiveness of these filters. Moreover, the oxidized As(V) is easier to remove when precipitated with Fe(III) or adsorbed onto the surface of an adsorbent. In research conducted by Nguyen et al. [14], Nguyen et al. [15], Petrushevski et al. [92], and Pradeep [17], oxidation and precipitation were evident in the filtration process. Normally, chemical oxidants such as O_3 , NaClO , and KMnO_4 can be used to effectively oxidize As(III). However, chemical oxidants are expensive and can produce undesirable by-products that demand extra treatment, raising the cost and complexity of the treatment. Consequently, given the limitations of oxidation chemicals, new ideas are produced through practical application.

In an experiment by Nguyen et al. [15], an aeration tank was designed to be included in a filter system, and its function was to facilitate the oxidation process of As(III) to As(V) and that of Fe(II) to Fe(III), to simulate the co-precipitation process between iron and As, and to remove the Fe–As precipitates. The oxidation processes of Fe^{2+} into Fe^{3+} (Equation (1)) and that of As(III) to As(V) (Equation (2)) can occur spontaneously when groundwater is extracted from a well and comes into contact with the oxygen in the air through the aeration tank. In this step, the As concentration fell significantly due neither to the co-precipitation process between Fe^{3+} (oxidized) and As(V), nor that between Fe^{2+} (un-oxidized) and As(V), which are demonstrated in Equations (3) and (4), respectively. This indicates that the aeration tank is capable of simulating the co-precipitation process between As and iron to produce the precipitates $\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$ and $\text{Fe}_3(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$. As with the aeration tank, the already existing household sand filters were used in a helpful pre-treatment step to improve the removal effectiveness and lengthen the timeline of the NLTT filter [15]. This meant that the groundwater was pre-treated with the traditional sand filter before passing through the NLTT filter. According to [15], sand filters are essential for the effective removal of As, Fe, and Mn from groundwater. The Fe/As ratio and oxidation conditions are, however, very important for this co-precipitation process, i.e., contact time and oxygen concentration. To successfully remove As from groundwater via co-precipitation, Meng et al. [100] recently found that an Fe/As ratio of less than 40 was required.



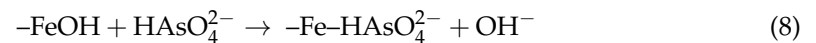
The oxidation process observed in ZVI adsorbent-based filtration systems is similar to the normal process. The mechanism by which ZVI adsorbents oxidize Fe(0) to Fe(II) and Fe(III) with oxygen—already present in water and in natural air—is fundamentally different from that of other iron-based adsorbents. The oxidation processes are represented in Equations (5) and (6). During the corrosion process, Fe(II) is produced by releasing the iron on the surface of the ZVI and the already existing partially dissolved Fe(II) in the filter column (Equation (5)). Moreover, a small amount of Fe(II) either re-adsorbs on the surface of the iron filings and the sand, or is never released from the iron surface at all. In aerated

water, Fe(II) that has been initially dissolved and Fe(II) that is adsorbed are both oxidized to Fe(III) (Equation (6)).



6.2.2. Adsorption

Following the oxidation process, the majority of the remaining As in the water, in the form of As(V), is removed utilizing adsorption on the Fe oxides/hydroxides via inner-sphere complexation. When iron-based adsorbents are exposed to water, their surfaces produce hydroxyl groups ($-\text{OH}_2^+$, $-\text{OH}^-$, and $-\text{O}^-$) that control their adsorption behavior. As can be removed via ligand exchange with hydroxyl groups on the surfaces of iron-based adsorbents, which forms an inner-sphere complexation. This process has been observed by Mohamed et al. [9], Siddiqui and Chaudhry [20], Mohan and Pittman [27], and Lakshmipathiraj et al. [101]. Equations (7) and (8) represent the reactions.



According to Lakshmipathiraj et al. [101], Goldberg and Johnston [102], and Jia et al. [103], the process by which arsenic is adsorbed onto iron-based adsorbents involves the formation of complexes because of the existence of numerous functional groups on the adsorbent's surface. Moreover, As can adhere to the iron-based adsorbent's surface either through physical adsorption or a chemisorption process.

Arsenic (As(III) and As(V)) is adsorbed onto iron-based adsorbents both through physical and chemical mechanisms. Chemical adsorption implies a chemical bond between a specific adsorption site on the adsorbent and the adsorbate. For example, electrostatic attraction is one of the most common chemical adsorption mechanisms by which iron-based adsorbents remove As. On the other hand, in the physical adsorption process, weak chemical interactions such as van der Waals interactions and hydrogen bonding occur between adsorbent and adsorbate. Alternatively, adsorption may occur in the microporous materials, a process called the pore-filling mechanism.

6.3. Cost of Filtration Systems

The field tests were predominantly carried out in Bangladesh, India, and Vietnam, nations whose ground water sources are often highly contaminated with As. According to the World Bank's assessment, these severely impacted nations are all categorized as low- and middle-income nations. Hence, one of the key determinants of successful filtration systems (along with other criteria, including removal efficiency, design, management of waste disposal, etc.) is cost. The high cost of certain filtration systems is a factor in the reduced number of As-removal techniques used in underdeveloped nations. The application of low-cost As treatment facilities is a key strategy that not only leads to the provision of safe As-free water sources in middle- and low-income countries, but also leads to millions of dollars in savings for these countries.

As is shown in Table 7, the cost of treating one cubic meter of water ranges from nearly nothing to USD 0.71. The cost of community filtration systems was reported to be USD 0.2/m³ for filter-based treated laterite, USD 0.09/m³ for filter-based natural laterite, USD 0.106/m³ for filter-based iron-coated sand, USD 0.61/m³ for filter-based confined metastable 2-line ferrihydrite, and USD 0.04/m³ for filter-based ZVI. Of these, the most inexpensive system used ZVI because this adsorbent can provide safe water with an As content <10 µg/L, making it an acceptable technology for India's rural areas. Moreover, the reported costs of household filtration systems using this same adsorbent were higher than that of the community filtration system. For instance, the cost per cubic meter of a

household filter incorporating treated laterite amounted to USD 0.35, which was slightly more costly than community filtration (USD 0.2). This trend agrees with the findings of Nguyen et al. [14], Nguyen et al. [15], and Pradeep [17]. Clearly, the cost estimates for each facility can be calculated using different variables. Some publications only provided the price per unit of treated water, while others specified capital and maintenance costs. Some published studies clearly reported the total investment and maintenance costs, while others just provided the price per unit of treated water.

Table 7. Practical application of iron-based adsorbents for removing As from groundwater.

Applied Adsorbent	Scale	Capacity	Location	Cost (USD/m ³)	As Conc. (µg/L)		Ref.
					Before Treatment	After Treatment	
Treated laterite	Community	500–1500 L/day	India	0.2	50–500	<10	[13]
Treated laterite	Household	100–120 L/day	India	0.35			[13]
Natural laterite	Household	10 L/h	Vietnam	0.71	122–237	1–10	[15]
Natural laterite	Community	500 L/h	Vietnam	0.09	27–235	0.4–10	[14]
Confined metastable 2-line ferrihydrite	Community	100–1000 L/h (gravity flow) 17 m ³ /h (electrically powered flow)	India	0.61	Up to 500	<10	[17]
Confined metastable 2-line ferrihydrite	Household	3 L/h	India	0.61–0.85			[17]
Iron oxide-coated sand (IOCS)	Community	55 m ³ /day	Hungary	0.106	16–285	<10	[91]
Iron oxide-coated sand	Household	100 L/day	Bangladesh	–	225–500	<10	[16, 92]
ZVI	Household, SONO filter	20–30 L/h	Bangladesh	–	140–960	<10	[94, 97]
ZVI	Household	10 L/h	Burkina Faso	–	400–1350	<50	[12]
ZVI	Household, Kanchan filter	15–30 L/h	Nepal	–			[95]
ZVI	Community	600–1000 L/h	India	0.04	60–400	<10	[93]
ZVI	Communities	31 L/day	Bangladesh	–	10–299	4–24	[104]
Activated alumina	Community	10–12 L/min	India	0.64	140	<50	[98]
Activated alumina	Community	8–10 L/min	India	–	100–850	<10–800	[99]

7. Conclusions

Iron-based adsorbents have gained considerable attention as potential tools for the removal of As from water environments due to their high efficiency, wide availability, and high affinity with As. In the literature, popular iron-based adsorbents are categorized into the following six groups depending on their components: iron oxides and hydroxides, iron-coated natural mineral adsorbents, iron-coated bio-adsorbents, iron-coated activated carbon, nanoscale zero-valent iron, and iron-based layered double hydroxides. Hundreds of these adsorbents have been studied in the laboratory for their ability to remove As from aquatic environments. However, there have been relatively few practical applications of these adsorbents. A few studies have applied iron-based adsorbents in community and/or household filtration systems to produce As-free water. The principle As-removal mechanisms of the iron-based adsorbents include oxidation, precipitation, and adsorption. The field tests were predominantly carried out in low- and middle-income countries such as Bangladesh, India, and Vietnam, where the groundwater is highly contaminated with As. Thus, cost is a crucial factor in determining the popularity of filtering systems, along with other criteria such as removal effectiveness, design, and waste management. The practical applicability of iron-based adsorbents should be further studied in the future with

a focus not only on As-removal performance, but also on investment costs, maintenance costs, As waste, disposal methods for exhausted adsorbents, and community awareness and participation.

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