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Review

Current Trends of Arsenic Adsorption in Continuous Mode: Literature Review and Future Perspectives

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Abstract: Arsenic is a toxic element for humans and a major pollutant in drinking water. Natural and anthropogenic sources can release As into water bodies. The countries with the greatest arsenic contamination issues lack the affordable technology to attain the maximum permitted concentrations. Adsorption can be a highly efficient and low-cost option for advanced water treatment, and the development of new cheap adsorbents is essential to expand access to water with a safe concentration of arsenic. This paper aims to review the state of the art of arsenic adsorption from water in continuous mode and the latest progress in the regeneration and recovery of arsenic. The disposal of the exhausted bed is also discussed. Fixed-bed column tests conducted with novel adsorbents like binary metal oxides and biosorbents achieved the highest adsorption capacities of 28.95 mg/g and 74.8 mg/g, respectively. Iron-coated materials presented the best results compared to adsorbents under other treatments. High recovery rates of 99% and several cycles of bed regeneration were achieved, which can aggregate economic value for the process. Overall, further pilot-scale research is recommended to evaluate the feasibility of novel adsorbents for industrial purposes.

Keywords: arsenic; adsorption; fixed-bed column; regeneration; biosorbents

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1. Introduction

Arsenic (As) is a metalloid from the pnictogen group of the periodic table, which can be released into water bodies from both natural and anthropogenic sources. Arsenic appears in water mainly from a combination of natural processes, such as weathering reactions, biological activity, volcanic emissions, and fluvial transport of As-containing mineral deposits [1,2]. In addition to natural causes, anthropogenic activities such as the use of arsenic compounds in agriculture and livestock, as well as mining and industry, are strong causes of groundwater and surface water arsenic contamination [3]. The prevention of arsenic water pollution from anthropological activities requires a proper water management plan and actions from local authorities; however, controlling natural sources of As is more complex and relies upon mitigation technologies and strict management of prevention measures [4].

Arsenic is a class 1 carcinogen, and human contamination can occur via consumption of contaminated water or food grown in contaminated soil. The most common inorganic forms are trivalent—As(III) and pentavalent—As(V), being the trivalent form much more toxic to human health [5]. The World Health Organization (WHO) and the European Directive 98/83/EC have established a guideline value of 10 µg/L for As in drinking water.

Consumption of contaminated water with As is a major concern in developing countries, which, in most cases, lack affordable technologies to attain WHO guidelines and EU parametric value. Among various existing techniques, adsorption is the most inexpensive and efficient process for advanced water and wastewater treatment [6]. Commercially available adsorbents, such as activated carbon, present some disadvantages, such as high production cost and difficulty in regeneration, leading to an increase in treatment costs [7].

Worldwide studies are being conducted to develop more efficient, economic, and sustainable adsorbents to remove arsenic from water, with biosorbents being one of the most promising alternatives to remove toxic metals and metalloids from contaminated waters.

Coating a common and cheap material to turn it into an adsorbent is a common practice among researchers to improve adsorption capacity and increase adsorption rate. The most common coating materials are iron, alumina, and other metals, such as zirconium and manganese. Beyond the increase in adsorbent capacity, iron-impregnated adsorbents are non-toxic, low cost, and accessible in large quantities [8].

Some review articles [8–11] have gathered the research into arsenic adsorption, but most of the studies discussed were conducted in batch mode, where the influent is treated sequentially (in cycles) in the reactor for a pre-determined time [12]. In continuous mode, the influent and effluent are continuously pumped into and out of the reactor. Figure 1 illustrates batch and continuous operation modes.

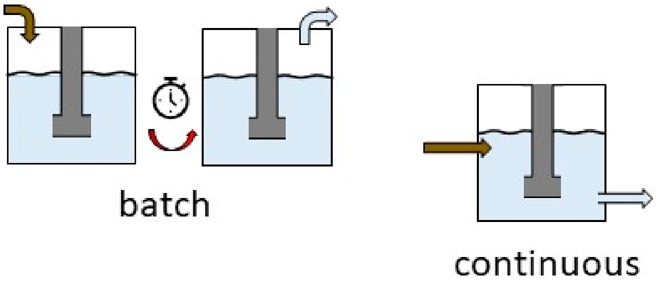

Figure 1. Illustration of batch and continuous treatment.

Adsorption in continuous mode is preferable for industrial and commercial purposes [6], hence the importance of investigating the state of the art of arsenic adsorption studies conducted in continuous mode, most commonly in a fixed-bed column.

This review article aims to investigate the tendency of arsenic removal on continuous mode, based on an analysis of published articles in the last 20 years, and also the progress in regeneration of exhausted bed and recovery of arsenic.

2. Methodology

In this article, a scoping literature review was carried out to gather studies from the last 20 years about arsenic removal from water in continuous adsorption processes. A total of 145 articles were gathered, with 33% being from the last 5 years (2016–2020), 15% from 2011–2015, 38% from 2006–2010, and 14% from 2000–2005. The main database used in this research was ScienceDirect, followed by Springer and Google Scholar. EndNote X9 was the software used to manage references.

The majority of selected articles are related to experimental studies of adsorption in continuous mode to remove arsenic from water. A few other articles were selected to provide complementary information. The studies were grouped by adsorbent type: metal (hydr)oxides (Section 3.1), zero-valent iron (ZVI) (Section 3.2), minerals (Section 3.3), soil and rock (Section 3.4), carbon-based adsorbents (Section 3.5), biosorbents (Section 3.6), industrial waste and by-products (Section 3.7), nanocomposites (Section 3.8), granular ferric hydroxide (GFH) and commercial adsorbents (Section 3.9), and layered double hydroxides (LDH) (Section 3.10). Figure 2 illustrates the types of adsorbents cited in this article.

Figure 2. Diagram of adsorbents types in As continuous adsorption studies.

From the selected literature, information about the recovery of arsenic and the regeneration of exhausted beds is summarized in Section 4. Similarly, information about the fate of contaminated beds is summarized in Section 5.

3. Adsorption of Arsenic in Continuous Mode: Adsorbents Type

3.1. Metal (hydr)oxides

Iron oxides are created by the precipitation of Fe^{3+}, and the resulting substance characteristics such as chemical structure, composition, and physical aspects can vary with different preparative techniques [13]. Iron oxides and hydroxides are the most common coating substances in sand columns for arsenic removal [14–20] due to their affinity with arsenic and relatively higher abundance in nature.

The combination of iron with another metal oxide is commonly investigated in column operation mode to increase arsenic removal [12,21]. Iron-zirconium binary oxide-coated sand (IZBOCS) achieved a maximum adsorption capacity of 28.95 mg/g by the Thomas model [15], while zirconium oxide-coated sand only achieved an adsorption capacity of 0.042 mg/g using the same adsorption model [22].

Dynamic tests with other metal-based adsorbents also show good results. Zirconium metal–organic framework column experiments were able to reduce arsenic initial concentration from 100 μg/L to 10 μg/L, for 2270 and 1775 bed volumes (BV) for As(III) and As(V), respectively [23]. Manganese performance was also investigated in column experiments conducted with natural manganese oxides [24] and manganese-coated sand (MCS) [14] for arsenic removal.

Table 1 shows characteristics of dynamic adsorption studies conducted with metal (hydr)oxides.

Table 1. Comparison between dynamic tests using metal (hydr)oxides and zero-valent iron (ZVI) as adsorbents for arsenic removal.

Adsorbent	Arsenic Species	Influent Concentration (µg/L)	Max. Adsorption Capacity (mg/g)	BV to Breakthrough Point of 10 µg/L	Breakthrough Time at 10 µg/L (h)	Ref.
Manganese-coated sand (MCS)	As	1000	0.079	250 pore volumes	18 days [1]	[14]
Iron-Zirconium Binary Oxide-Coated Sand (IZBOCS)	As(V)	125	25.09–28.95		0.5–0.75	[15]
Natural iron mineral-quartz sand columns	As(V)	500		7000 pore volumes		[16]
Crystalline hydrous ferric oxide (CHFO)	As	320–400		14,000		[20]
Zirconium oxide-coated sand	As(III)	500	0.03310–0.04223		5.33–8.33	[22]
Zirconium metal organic frameworks (UiO-66)	As(III)	100		2270		[23]
	As(V)	100		1775		[23]
Hydrated stannic oxide (HSO)	As(III)	1000		2400		[25]
	As(V)	1000		450		[25]
Zero valent iron (ZVI)	As(V)	100		1900		[26]
				500–12,600 PV [2]		[27]
					20	[28]

[1] Pilot-scale [2] For a breakthrough point of 1 µg/L.

The hybrid adsorbent of iron and zirconium (IZBOCS) achieved the highest value of adsorption capacity of 28.95 mg/g. The crystalline hydrous ferric oxide (CHFO) was able to treat a great amount of arsenic-contaminated solution (14,000 BV) with an outlet concentration below the WHO recommended value. The longer time of breakthrough of 18 days was achieved by manganese-coated sand (MCS) in a pilot-scale plant. Metal (hydr)oxides are usually impregnated in other materials to increase arsenic removal efficiency or/and reduce the treatment costs, as will be discussed in the following sections.

3.2. Zero-Valent Iron (ZVI)

The mechanism and behavior of arsenic adsorption by zero-valent iron (ZVI) in dynamic systems was investigated in some studies [26–31]. Earlier investigations in column mode with zero-valent iron filings, which were carried out with an inlet arsenic solution of 100 µg/L (pH 7.0), led to an outlet concentration below 1 µg/L for 500 and 12,600 pore volumes (PV), with a time residence of 20 s and 25 min, respectively [27].

Results of dynamic adsorption studies conducted with zero-valent iron are shown in Table 1. ZVI presented a performance similar to metal (hydr)oxides, however, the column studies with ZVI showed a substantial iron leaching of 8 to 17 mg/L in the final effluent [28,29], which can be a hindrance in the application of ZVI to remediate contaminated waters. The corrosion of iron by acid waters increases the iron release, which can result in an effluent with an iron concentration of 90–130 mg/L [28].

3.3. Minerals

Column experiments to investigate arsenic removal were carried out using a variety of minerals as adsorbents: akaganeite [32], attapulgite [33], bauxite [34–37], goethite [38], hematite [16,39], mackinawite [40], magnetite [41–44], siderite [16,39,45], schwertmannite [46], and zeolite [47,48]. Iron and aluminum-based minerals are considered good adsorbents due to their known affinity for heavy metals and metalloids and due to their abundance in nature.

Iron modified calcined bauxite (MCB) was investigated in column experiments for arsenic removal from aqueous solution [34–37]. The breakthrough point of 10 µg/L for a 10 cm bed depth (2 cm diameter) was reached after 31 h (~270 BV) at 5 mL/min with an As(V) solution of 2 mg/L [34]. The breakthrough point (10 µg/L) for an As(III) solution of 1 mg/L, in similar conditions of flow rate and bed depth, was reached after 50 h (~470 BV) [36]. The MCB maximum adsorption capacities reported in column mode were 0.606 mg/g and 0.490 mg/g for As(V) and As(III), respectively [34,36].

A novel adsorbent consisting of magnetite-rich particles (MEP) was developed with a low magnetic intensity separation from mill scale, which is constituted of wustite (FeO), hematite (α-Fe_2O_3), magnetite (Fe_3O_4), elemental iron, and residual oil and grease [43]. Column adsorption tests were carried out in four sets of packed-bed columns, each with 3.5 cm diameter, 20 cm bed height and 550 g of MEP. A synthetic wastewater containing 500 µg/L of As(V) was fed into the column at different flow rates. Breakthrough point (~10 µg/L) was reached after about 17 d and 30 d, at a feed flow rate of 6.4 mL/min and 3.2 mL/min, respectively. After saturation, at the 48th d of operation, MEP columns were regenerated with 0.2 N NaOH for 24 h at a flow rate of 1 mL/min, and the sorption capacity was reinstated to the initial condition.

Column tests with naturally occurring siderite, an iron carbonated mineral, led to a final effluent below 1.0 µg/L (26,000 pore volumes) using a solution containing 250 µg/L As(III) and 250 µg/L As(V) [45]. A similar study with a column packed with half siderite and half hematite content achieved As concentration of 1.0 µg/L after 7200 pore volumes (PV) [45]. The addition of hematite in siderite filters reduces the arsenic removal efficiency because siderite is usually more efficient to uptake As from water than hematite [49]. Siderite-hematite filters were activated with H_2O_2 which increased arsenic adsorption efficiency up to a throughput of 500 PV [39].

Three types of granular schwertmannite, an Fe(III)-oxyhydroxy sulfate mineral, were investigated in column tests for arsenate removal [46]. The experiments were conducted in a lab-scale column with 2 cm diameter and 20 cm bed height, fed with simulated groundwater containing 210 µg/L of As(V). The irregular shape (IS) type achieved the best performance and breakthrough of 10 µg/L occurred after 8100 BV, followed by the cylindrical shape (CS) type with 4200 BV and the spherical shape (SS) type with 120 BV. The adsorption capacities of IS, CS, and SS were 0.93, 0.66, and 0.33 mg/g, respectively.

Table 2 shows the results of dynamic adsorption studies conducted with minerals. It is possible to highlight the performance of magnetite derived from mill scale compared to other column studies using minerals such as arsenic adsorbent. Natural siderite also achieved good results, even better than the siderite-hematite media. The other minerals, most of them modified iron, presented relatively similar results for arsenic removal.

Table 2. Comparison of dynamic tests using minerals as adsorbents for arsenic removal.

Adsorbent	Arsenic Species	Influent Concentration (µg/L)	Max. Adsorption Capacity (mg/g)	BV to Breakthrough Point of 10 µg/L	Breakthrough Time at 10 µg/L (h)	Ref.
Iron modified calcined bauxite (MCB)	As(V)	2000–8000	0.470–0.606	269.2–300.64	2.15–59	[34]
Iron modified calcined bauxite (MCB)	As(III)	1000		428.02–489.17	28–96	[35]
Iron modified calcined bauxite (MCB)	As(III)	1000	0.392–0.459	427.85–489.17	28–96	[36]
Iron-modified clinoptilolite	As(V)	100		300		[47]

Table 2. *Cont.*

Adsorbent	Arsenic Species	Influent Concentration (µg/L)	Max. Adsorption Capacity (mg/g)	BV to Breakthrough Point of 10 µg/L	Breakthrough Time at 10 µg/L (h)	Ref.
Mill-scale derived magnetite particles	As(V)	100	3.60–5.00		>100	[42]
Magnetite-enriched particles (MEP)	As(V)	500			17–30 days	[43]
Mixed-valent iron oxide/magnetite	As(V)	100–1000	10		888–1032	[44]
Natural siderite	As(III) + As(V)	250 + 250	1.090–2.000	11,600–26,000 PV		[45]
Siderite-hematite	As(III) + As(V)	250 + 250		7200 PV		[45]
Activated siderite-hematite	Total As	500	0.177–0.185	8160 PV		[39]
Goethite-polyacrylamide composite (goethite-P(AAm)	As	300		400 [1]		[38]
Granular schwertmannite	As(V)	200	0.33–0.93	120–8100		[46]
Iron-coated zeolite (ICZ)	As(V)	2000	0.69	300		[48]
Iron impregnated charred granulated attapulgite	As(V) As(III)	200		397 175		[33]

[1] For breakthrough point of 25 µg/L.

3.4. Soil and Rock

Soils and rocks are also natural abundant materials, often containing minerals that enhance their arsenic uptake capacity. They stand out from other adsorbents due to their advantages of being low-cost and locally available.

The arsenic uptake capacity of natural occurring laterite soil was studied in column operation mode using arsenic solutions and real samples [50–53]. Fixed-bed column studies conducted in a 2 cm diameter column and a bed depth of 30 cm, fed with arsenic solution of 500 µg/L, at 7.75 mL/min flow rate, reached the breakthrough point at 10 µg/L after 32 h for As (V) [52] and 18.5 h for As (III) [51]. The total arsenic removal by laterite soil, in similar column experiments (10 cm bed depth), with a groundwater (tube-well) sample of 0.33 ppm of arsenic was 98% [50].

Moreover, recent research has developed a novel laterite soil filter for arsenic that provides potable water to a population higher than 5000 people [54]. A model to predict the lifetime of the filter was validated by lab-scale and field-scale experiments. It was predicted that the novel laterite filter could last from 10 to 100 months, depending on flow rate, size, and the arsenic concentration of the feed water.

The arsenic adsorption behavior of another soil, originated from limestone and heavily iron-coated, was investigated [55] in a glass column with 1 cm diameter, packed with 2 g of adsorbent (1.7 cm depth). The system was fed with an arsenate solution of 1 mg/L, in order to investigate the effect of pH, pore water velocity, and the presence of phosphate (PO_4^{3-}). The phosphate ions produced a major effect on As(V) mobility and recovery, followed by the pore water velocity effect, with the least effect being observed due to pH variation.

A few studies reported rocks as adsorbents for arsenic removal from water. Pisolite, a mining waste composed of iron and manganese aggregate, was applied for this purpose [56]. After 180 min of column operation, 1.0 g of pisolite removed 1.41 mg of As, and 1.0 g of activated (400 °C heating) pisolite removed 3.51 mg of As. Native limestones were investigated in column experiments fed by solutions with similar characteristics to As-rich well water from Mexico [57]. The authors observed the interference of bicarbonate and sulfate in As removal and indicated native limestones for the treatment of arsenic-contaminated water. Diatomite coated with hydrous ferric oxide (HFO) was able to treat about 1100 BV of As(III) solution (500 µg/L) until a breakthrough point of 10 µg/L [58].

The arsenic removal efficiency by an iron-oxide-coated natural rock (IOCNR) was studied in a fixed-bed column with 2 cm diameter, fed at an 8 mL/min flow rate at a pH of 5.7 [59]. The rock used in the study was Fe(III)-loaded granite. The breakthrough point of 10 µg/L for an initial As(III) concentration of 600 µg/L was reached after 31 h, 49 h, and 63 h for bed depths of 10 cm (~48 g), 15 cm (~73 g), and 20 cm (~97 g), respectively. A similar dynamic study was conducted with IOCNR, and the adsorption-exhausted sludge of As(III) and As(V) was mixed with Portland cement to reduce the impact and the cost of the water treatment process [60]. The study evaluated the As mobility in the material, and the leaching test allowed the authors to conclude that the adsorption column sludge mixed with Portland cement is an environmentally friendly material that could be applied in building construction.

Table 3 shows characteristics of dynamic adsorption studies conducted with soil and rock.

Table 3. Comparison of dynamic tests using soil and rock as adsorbents for arsenic removal.

Adsorbent	Treatment	Arsenic Species	Influent Concentration (µg/L)	Breakthrough Time at 10 µg/L (h)	Max. Adsorption Capacity (mg/g)	BV to Breakthrough Point of 10 µg/L	Ref.
Laterite soil	None	As	330	6.75			[50]
Laterite soil	None	As(III)	500–1000	2.8–32			[51]
Laterite soil	None	As(V)	500–1000	1.65–18.5	0.047		[52]
Laterite soil	None	As(V)	500–1002	4.95–8.12	0.112–0.147		[53]
Laterite soil	None	As	80–100				[54]
Iron-oxide containing soil	None	As(V)	1000			15–875	[55]
Pisolite	Thermic (400 °C)	As(V)	50,000		1.41–3.51		[56]
Limestones	None	As(V)	1190–1340	3–15 weeks			[57]
Iron-incorporated diatomite [Fe(25%)-diatomite)]	$Fe(NO_3)_3 \cdot 9H_2O$	As(V) As(III)	500 500			1100 1100	[58]
Iron-oxide-coated natural rock (IOCNR)	$Fe(NO_3)_3 \cdot 9H_2O$	As(III)	600	31–63			[59]
		As(III) As(V)	300–1000 1000–3000	20–65 5–18			[60]
Iron-impregnated tablet ceramic (ITCA)	$FeCl_3$	As(V)	1000	16			[61]

Rock-based adsorbents presented better results for arsenic removal than soil-based ones. The longest breakthrough time was achieved by limestones, which maintained arsenic effluent solution under 10 µg/L for weeks, followed by IOCNR, which kept effluent concentration under the WHO standard for more than 60 h.

3.5. Carbon-Based Adsorbents

Most of the carbon-based adsorbent investigations in column operation mode for arsenic adsorption have been conducted with activated carbon (AC) or granular activated carbon (GAC) loaded with iron to benefit from the properties of both materials [62–67]. The use of AC, especially GAC, as a support for metal oxides/hydroxides, provides a more stable structure for As removal in comparison with the application of the metal oxide/hydroxide alone, mainly in fixed-bed column processes [63].

A continuous mode study using iron-impregnated GAC (GAC-Fe) and pristine GAC was conducted for As(V) removal [63]. The maximum As adsorption capacity observed was 0.470 mg/g for the GAC-Fe column, which performed approximately four times better than the GAC column of 0.133 mg/g [63].

Hydrous ferric oxide was immobilized onto GAC using phenol formaldehyde (PF) to provide an economically feasible solution for As removal from water [66].

Another methodology to prepare ferric AC was by precipitating amorphous HFO onto AC (FeO/AC) [65]. Laboratory column experiments with FeO/AC were performed at 8.33 mL/min with an As(V) initial concentration of 500 µg/L. FeO/AC was able to remove at least 99.8% of arsenic from the influent, and the breakthrough point of 50 µg/L was achieved after approximately four times more throughput volume compared to AC without iron loading. Table 4 presents the results of dynamic adsorption studies using carbon-based adsorbents. The iron impregnation of GAC and AC was able to increase As uptake from water by about four times, considering results of column experiments [63,65]. However, GAC/AC are usually more expensive adsorbents, and their adsorption capacities obtained in dynamic experiments were found to be similar or inferior to other low-cost adsorbents such as minerals, soil and rock, biosorbents, and industrial waste and by-products.

Table 4. Comparison of dynamic tests using carbon-based adsorbents for arsenic removal.

Adsorbent	**Treatment**	**Arsenic Species**	**Influent Concentration (µg/L)**	**Max. Adsorption Capacity (mg/g)**	**BV to Breakthrough Point of 10 µg/L**	**Breakthrough Time at 10 µg/L (h)**	**Ref.**
Iron-modified activated carbon	$Fe(NO_3)_3 \cdot 9H_2O$	As	40–60		5600–34,000		[62]
Iron-impregnated granular activated carbon (GAC-Fe)	$FeCl_2$	As	100–500	0.305–0.470	313–1494		[63]
Granular activated carbon (GAC)		As	100	0.118–0.133	380–388		[63]
Granular activated carbon doped with iron (Fe/GAC)	$FeCl_3$	As(V)	180	2.873			[64]
Hydrous ferric oxide incorporated onto GAC with phenol formaldehyde resins coating (HFO-PF-coated GAC)	$FeCl_3$/PF	As(V)	1886	0.7117	180	1753	[66]
Iron-containing granular activated carbon	$FeCl_2$	As(V) As(III)	57.2 56.1		7500 7500		[67]

3.6. Biosorbents

Biosorbents are naturally occurring materials or waste biomass, and their high adsorption capacity and cost-effectiveness make biosorption a promising process in water and wastewater treatment [68]. The group of biomaterials includes aquatic biopolymers, plant biomass, microbial biomass, biochars, and agricultural waste [69]. Column studies have investigated the potential of these materials, with or without pre-treatment or modification, to remove arsenic from aqueous solution in continuous mode [7,70–97].

3.6.1. Aquatic Biomaterials

Chitosan

Chitosan (CS) is a natural aminopolysaccharide originating from exoskeletons of arthropods [7] such as crabs and shrimps. The biopolymer is obtained by *N*-deacetylation of chitin [78] and has been applied in heavy metals and arsenic uptake due to its free amino groups that can serve as sites of coordination binding [72]. Chitosan also presents the advantage of being low cost, biodegradable, renewable, and non-toxic [75].

Early experimental column studies with chitosan beads investigated the feasibility of using chitosan without any pretreatment to remove arsenic from wastewater discharged from gallium arsenide (GaAs) manufacturing in continuous mode [72]. Laboratory tests were developed in a glass column with 1.8 cm diameter and 12 cm height with a total packing volume of 30.5 mL, packed with 20 g of chitosan beads of 2.5 mm size. The industrial wastewater contained 2.5 mg/L of As(III) and 35 mg/L of As(V) and presented a pH of 4.8. Comparison tests were made with synthetic wastewater containing 10 mg/L of As(III) or As(V), at pH 5. Recycled chitosan beads were also investigated in continuous mode and presented similar breakthrough time to native chitosan beads. The experiments made with synthetic wastewater at 8 mL/min resulted in an adsorption capacity of 1.78 and 1.90 mg As/g bead, for As(III) and As(V), respectively. According to this study, chitosan is a promising adsorbent for arsenic removal from water.

Later, molybdate-impregnated chitosan beads (MICB) were developed to improve arsenic adsorption capacity [73]. Laboratory column tests were carried out with synthetic effluent and GaAs manufacturing wastewater with similar characteristics to the previous study [72]. The breakthrough time at 30 mL/min was approximately 7 h for the synthetic wastewater and between 5 h and 8 h for the industrial wastewater. Recycling tests were made, and the adsorption curve remained similar up to 10 cycles of sorption–desorption. MICB adsorption capacity observed with synthetic wastewater was 1.98 and 2.00 mg As/g MICB for As(III) and As(V), respectively. The adsorption capacities observed for MICB were only slightly higher than the ones observed for chitosan without pretreatment, so the costs of adding molybdate were not justified by an incrementation in the adsorption capacity.

In addition to molybdate impregnation for chitosan beads, studies have proposed other metal or inorganic oxide treatments to improve their arsenic adsorption capacity. Iron oxides are the most common [7,74–76,78,83], followed by aluminum oxides [70,74], magnesium oxide [81], and the combination of iron and manganese oxides [7].

Among the column studies of chitosan treated with metal oxides, the best arsenic adsorption capacity was observed for chitosan entrapped with Fe-Mn water treatment residual (C-WTR), with an As(III) removal capacity of 32.64 mg/g of C-WTR even after six cycles of sorption/desorption with 0.5 NaOH solution [78].

In addition to the treatment of chitosan with metal oxides, a simple method of protonation of amine groups, improved chitosan arsenic adsorption capacity on column tests to a range of 36–50 mg As/g of chitosan [71,80], which is the highest adsorption capacities among other types of chitosan modifications reported in column studies.

Alginate

Alginate is a biopolymer composed of α-l-guluronate and β-d-mannuronate and is a major constituent of the cell wall of seaweed and sea tangle [77,98]. Alginate acids have high affinity with divalent cations and produce thermally irreversible and hydrophobic gels [84,99]. Alginate beads are applied to heavy metals and metalloids removal, and their preparation is usually made with calcium as cation [84]. Arsenic adsorption column studies using alginate treated with metal oxides, such as aluminum oxide [77] and iron oxides [82,84,100] have been reported.

An alginate-based adsorbent containing calcinated alum sludge from a water treatment facility was developed to enhance surface area and overcome alginate bead limitation of slow adsorption kinetics [77]. The new adsorbent performed better than a commercial adsorbent (Bayoxide E33) by reaching 10 μg/L after 200 bed volumes, 50% higher than Bayoxide E33. The initial solution concentration was 200 μg/L.

Fe(III) crosslinked alginate nanoparticles were also investigated in fixed-bed column experiments for As(V) removal [101]. The effect of inlet arsenate concentration (0.5, 1.0, and 1.5 mg/L), bed height (2, 4, and 6 cm) and flow rate (0.25, 0.50, and 1.0 mL/min) were studied. The Fe-alginate maximum adsorption capacity of 0.066 mg/g was achieved with the following dynamic experimental conditions: inlet As(V) solution of 0.5 mg/L, bed height of 2 cm, and flow rate of 0.50 mL/min.

Fish Scale

The atlantic cod fish scale has been reported as an alternative biosorbent to remove heavy metals and arsenic from water [79,102,103]. Column tests were conducted using a dried fish scale in a glass column with 2.54 cm diameter and 23.88 cm bed height. The column was fed with a solution of 520 μg/L As(III) or 342 μg/L As(V) at a flow rate of 2.0 mL/min. The breakthrough point ($C/C_o = 0.1$) was reached after 21.75 h and 25 h for As(III) and As(V), respectively.

3.6.2. Agricultural Waste

Agricultural waste materials are mainly composed of lignin and cellulose, combined with other components such as hemicellulose, extractives, lipids, proteins, starches, water, hydrocarbons, and others [104]. This type of waste is a potential source of adsorbents for heavy metals and metalloids. The majority of research is directed to cationic heavy metals, and few studies have investigated the capacity of various food and agricultural wastes for the biosorption of arsenic [9].

Carbonized sugarcane bagasse (SCC) is an alternative to active char carbon prepared from agricultural waste material [94]. Column tests were performed to investigate the arsenate and arsenite removal capacities of SCC modified by thioglycolic acid impregnation [94]. Breakthrough curves were generated at different bed heights (11.3 cm to 32.1 cm), flowrates (3.0 to 7.0 mL/min), and arsenic initial concentration (500 to 1500 μg/L) with a pH of 6.0. Experimental and predicted (by Thomas model) adsorption capacities for As(III) and As(V) were found. Maximum adsorption capacities of 0.085 mg/g and 0.084 mg/g were reached for As(III) and As(V), respectively, at a flow rate of 3.0 mL/min, bed height of 32.1 cm and initial concentration of 1500 μg/L.

Rice-Based Adsorbents

Rice polish, a waste from rice milling industries, was investigated as a potential adsorbent in up-flow fixed-bed column studies [93]. The effect of design parameters, such as bed height (5 to 25 cm), flow rate (1.66 to 8.33 mL/min), and initial metal ion concentration (100 to 1000 μg/L), was studied. Maximum uptakes of 0.067 mg/g for As(III) and 0.079 mg/g for As(V) were achieved with 25 cm bed height and 30.5 g of rice polish, at 1.66 mL/min flow rate, fed with a 1000 μg/L initial arsenic concentration. Desorption–sorption cycles were also investigated, and the recovery efficiency achieved values of approximately 99%.

A good adsorption of arsenic performance was observed for rice waste studies in continuous mode [93,95]. Considering that rice production is strong in some places where arsenic contamination is a major problem, such as Bangladesh and other Asian countries, rice by-products can be a low-cost potential solution for arsenic removal from water in these regions.

Cotton-Based Adsorbents

Column studies with cotton-based adsorbents were performed after iron modification of the materials [91,97]. A novel adsorbent, Fe(III)-loaded ligand exchange cotton cellulose [Fe(III)LECCA] was synthetized for selective arsenate adsorption [97]. In column tests with 1 mg/L arsenic aqueous solution, at pH of 7.10, a breakthrough capacity (at 10 μg/L) of 5.3 mg/g was achieved with a flow rate of 26 BV/h. In similar column tests with tap water spiked with As(V), the breakthrough point adsorption capacity was reduced to 65% of the original value.

Fixed-bed column experiments were also performed with another novel adsorbent, consisting of Fe-based metal-organic framework coated onto cotton fiber composites [MIL-88A(Fe)/cotton fibers] [91]. A solution with an As(V) concentration of 500 μg/L was fed at 6.0 mL/min flow rate to the column with 34 cm length and 1.6 cm diameter. The breakthrough point of 10 μg/L was achieved after approximately 440 bed volumes.

Another cotton-based adsorbent study in column mode was reported, but instead of iron, zirconium was loaded to the material to improve uptake capacity [87]. The novel adsorbent was synthesized by radiation-induced graft polymerization (RIGP) of a phosphoric monomer onto nonwoven cotton fabric. Zr(IV) was then loaded to the phosphoric units packed in the column. The new Zr-type adsorbent arsenic removal capacity was investigated in column tests at different pH (1, 2, 3, 5, 7, and 9) and different flow rates (200 and 1000 h^{-1}) with an As(V) solution of 1 mg/L. The maximum adsorption capacity of 0.749 mg/g was obtained at pH 2 and 1000 h^{-1} flow rate.

Among the agricultural wastes used in dynamic studies for arsenic removal, the cotton-based materials led to better adsorption capacities because of modifications with iron and zirconium. Moreover, iron loading has provided higher adsorption capacity to cotton-based adsorbents than zirconium.

3.6.3. Biochar

Biochar is a stable black carbon material produced from a wide range of biomass types [105]. The biochar characteristics such as large specific surface area, porous structure, enriched surface functional groups, and mineral components, combined with its being environmentally friendly, low-cost, and effective, make the application of biochar as an adsorbent attractive to researchers [105,106]. Recent studies reported the association of iron composites to biochar in order to improve arsenic adsorption removal in column mode [41,88,96].

Biochar derived from hickory chips was impregnated with iron nanoparticles by direct hydrolysis of iron salt and investigated for arsenic adsorption–desorption [88]. A column measuring 5 mm in diameter and 12 mm of bed height was filled with 1 g of the iron-impregnated biochar and fed with a 50 mg/L As(V) solution at 2 mL/min. Column experimental data were well fitted by the convection-dispersion-reaction (CDER) model. Desorption tests carried out with 0.05 mol/L $NaHCO_3$ obtained an 85% removal of the retained arsenic.

Magnetized biochar composite (MBC) was prepared by precipitating magnetite primary nanoparticles (Fe_3O_4) on Douglas fir biochar [41]. Fixed-bed column experiments were performed using a 10 mg/L As (III) solution at pH 7 at 2 mL/min flow rate, and the breakthrough point was reached after 40 min. Regeneration tests were conducted with 0.5 M KH_2PO_4 at pH 4.5, in three cycles, and results show that restoration rates were 85%, 81%, and 76% from the initial adsorption capacity. The desorption efficiency was reduced from 87% to 80% and 76%.

Recently, iron oxide nanoneedles array-decorated biochar fibers (Fe-NN/BFs) were prepared by a hydrothermal method from waste cotton fiber [96]. Fixed-bed columns containing 2 g of adsorbent were fed with 275 μg/L arsenic spiked natural groundwater at 2.26 mL/min and at pH 6. For a breakthrough point of 10 µg/L, the treated volumes for As(V) and As(III) were 1350 and 350 bed volumes, respectively. For two columns in tandem, the treated volume increased to 2900 BV for As(V) and 2500 BV for As(III). The adsorption capacity of the column was 1.80 and 1.55 mg/g for As(V) and As(III), respectively. Five cycles of regeneration (0.1 M NaOH) were conducted with effective treatment performance.

3.6.4. Fungal Biomass

A recent study investigated the arsenic adsorption capacity of four novel fungal strains in column mode [89]. *Westerdykella* sp. (FNBR_3), *Trichoderma* sp. (FNBR_6), *Lasiodiplodia* sp. (FNBR_13), and *Rhizopus* sp. (FNBR_19) were isolated from agricultural soils containing arsenic in West Bengal, India. Column experiments were performed in a glass column (0.7 cm diameter and 15 cm bed height) packed with alginate beads containing 10 g of immobilized fungal biomass at pH 6 with a flow rate of 8.5 mL/min and arsenic initial concentration of 200 mg/L. *Lasiodiplodia* sp. (FNBR_13) led to the highest values of breakthrough time (Cb = 100 µg/L) and adsorption capacity: 110 min and 74.88 mg/g, respectively. The Thomas model was fitted to the experimental results and the highest correlation, 0.9799, was exhibited by FNBR_13 experimental data.

In another study, *Aspergillus niger* fungal biomass was treated with iron oxide to remove As(III) and As(V) in continuous mode [92]. A mass of 6.925 g of iron oxide-coated biomass (IOCB) beads were packed in a column with 1.25 cm diameter and 23.7 cm effective bed depth. Different solutions of 100 µg/L As(III) or As(V) at pH 6 were fed to the column at 3.1 mL/min flow rate. The breakthrough point of 10 µg/L for As(V) was achieved after 800 bed volumes for IOCB and after 130 bed volumes for regenerated IOCB (0.25 M NaOH at 10 mL/min). The As(III) breakthrough point could not be achieved in the first cycle, but it was achieved with regenerated IOCB after 340 bed volumes. The adsorption capacities of IOCB were reported to be 1.08 mg/g for As(V) and 1.21 mg/g for As(III).

Fungal biomass can be considered a good source of biosorbents for arsenic removal. However, high values of adsorption capacity reported in the first study [89] can be explained by the high arsenic initial concentration of 200 mg/L, 20,000 times higher than the initial arsenic solution in the second study [92]. The second study reported that fungal biomass with iron modification was able to achieve adsorption capacities in continuous mode similar to other biosorbents' removal capacity.

3.6.5. Plant Biomass

The arsenic adsorption capacity of sorghum biomass (SB), one of the greatest cereal crops in the world, was investigated in continuous mode [86]. Two columns with 5.4 cm diameter and 40 cm active bed height were fed with a solution containing 5 mg/L of As(III) at 10 mL/min. One column was packed with 150 g of non-immobilized sorghum biomass (NISB) and the other one with 140 g of immobilized sorghum biomass (ISB). The maximum adsorption capacities observed for NISB and ISB were, respectively, 2.765 and 2.437 mg/g.

Preliminary experiments were conducted with immobilized *Garcinia cambogia* biomass beads (IGCFIX) to remove As(III) in continuous-flow conditions [90]. A column with 1.6 cm diameter and 7 cm of packed bed height was fed with a 100 mg/L As(III) solution at a flow rate of 13.3 mL/min. A regeneration study was carried out with 0.05 M NaOH at 1 mL/min flow rate in five cycles. The results achieved point to the feasibility of using IGCFIX in column operation.

Table 5 shows characteristics of dynamic adsorption studies conducted with carbon-based biosorbents. The average performance of biosorbents in column studies was superior to that of traditional types of adsorbents, such as metal (hydr)oxides and ZVI (Table 1) minerals (Table 2), soils and rocks (Table 3), and carbon-based materials (Table 4). The different categories of biosorbents showed similar results but more studies were conducted with aquatic biomaterials, which can justify their higher feasibility to column studies.

Individually, the cod fish scale and chitosan-based materials showed higher maximum adsorption capacities than other biosorbents.

Table 5. Comparison of dynamic tests using biosorbents for arsenic removal.

Adsorbent	Arsenic Species	Influent Concentration (μg/L)	Breakthrough Time at 10 μg/L (h)	Max. Adsorption Capacity (mg/g)	BV to Breakthrough Point of 10 μg/L	Ref.
Aquatic Biomaterials						
Chitosan	As(V)	30,000–120,000	424.7–4444.6	36–50		[71]
Chitosan	As(V)	120,000	4.33			[80]
Chitosan	As(V) As(III)	10,000 10,000		1.9 1.78		[72]
Molybdate-impregnated chitosan (MICB)	As(V) As(III)	10,000 10,000		1.99 1.96		[73]
Chitosan-based porous magnesia-impregnated alumina (MIPA)	As(V)	300	42–100	17.2		[81]
Porous Fe-chitosan beads (P/Fe-CB)	As(III)	975		1.19	3000	[83]
Iron chitosan spacer granules (ICS)	As(V) As(III)	500–1000 500–1000	59.0–106.0 50.5–107.7		210 132	[76]
Waste Fe/Mn oxides into chitosan beads (C-WTR)	As(V) As(III) As(III) + As(V)	500 500 500 + 500		13.84–13.97 30.02–32.64	2700	[78]
Magnetic binary oxide particles (MBOP) using chitosan	As(III)	1000	26.15–72	0.436–0.477		[74]
Granular adsorbent (GA) using back-wash residue material and chitosan	As(V)	150			210	[7]
Iron oxide loaded alginate	As(V) As(III)	50		0.0138	230 45	[84]
Ferric hydroxidemicrocapsule loaded calcium alginate (FHMCA)	As(III)	100	75 days			[82]
Atlantic cod fish scale	As(V)	342–520	21.75–25	4.87–38.36		[79]
Agricultural Waste						
Iron(III)-loaded phosphorylated orange juice residue (POJR)	As(V)	15,800			81.4	[85]
Fe-based metal-organic framework/cotton fibers composites	As(III)	500			320–440	[91]
Rice polish	As(V) As(III)	100–10,000	3–30.83 2.66–29	0.025–0.079 0.030–0.067		[93]
Rice husk	As(III)	50–500		0.016–0.022		[95]

Table 5. *Cont.*

Adsorbent	Arsenic Species	Influent Concentration (µg/L)	Breakthrough Time at 10 µg/L (h)	Max. Adsorption Capacity (mg/g)	BV to Breakthrough Point of 10 µg/L	Ref.
Fe(III)-loaded ligand exchange cotton cellulose adsorbent [Fe(III)LECCA]	As(V)	1000		5.3	~400	[97]
Nonwoven cotton fabric	As(V)	1000		0.749	270–1420	[87]
Thioglycolated sugarcane carbon (TSCC)	As(V) As(III)	1500 1500		0.084 0.085		[94]
Biochar						
Iron-impregnated biochar	As	50,000	0.167			[88]
Magnetite precipitated onto Douglas fir biochar	As(III)	10,000		2.85		[41]
Iron oxide nanoneedle array-decorated biochar fibers (Fe-NN/BFs)	As(V) As(III)	275 275		1.80 1.55	1350 350	[96]
Fungal and Plant Biomass						
Non-immobilized sorghrum biomass (NISB)		5000	27 [1]	2.765	31 pore volumes [1]	[86]
Immobilized sorghum biomass (ISB)		5000	24 [1]	2.437	27 pore volumes [1]	[86]
Novel fungal strains + alginate beads	As	200,000	0.50–1.83 [2]	59.5–74.8		[89]
Iron oxide-coated fungal biomass (IOCB)	As(V) As(III)	100 100		1.080 1.210 [3]	800 340 [3]	[92]

[1] For breakthrough point of 50 µg/L [2] For breakthrough point of 100 µg/L [3] Regenerated bed.

3.7. Industrial Waste and By-Products

Looking for industrial waste materials for arsenic uptake and other pollutants' removal from water is of great importance to improve sustainability because it can be cost-effective and also reduce waste disposal in industry. Column experiments have shown good potential for arsenic adsorption using sugar industry waste [107], water treatment solid residuals [77,108], cement [109–111], mining residual products [57], and, more recently, waste and by-products from scrap metal recycling [112] and the steel industry [42–44].

Regarding sugar industry waste, column experiments for arsenic removal were developed using, as adsorbent, bagasse fly ash (BFA) [107]. The column data were analyzed by bed depth service time (BDST) and Yoon and Nelson models. The arsenate and arsenite removal efficiencies in synthetic influents were 98.9% and 95.6% and for real arsenic-contaminated water were 91.23% and 87.76%, respectively. The authors concluded that the BFA adsorption column is applicable to natural water remediation as regards As contamination.

Chemical sludge rich in iron from a water treatment plant (WTP) in the USA was also investigated as a potential adsorbent for arsenic [108]. The materials were rich in iron because the WTP used ferric sulfate as coagulant. Column tests were conducted using the ferric sludge and granular ferric hydroxide (GFH) for comparison purposes. The ferric residual presented a performance similar to GFH and achieved a breakthrough point of 10 µg/L after 26,400 bed volumes, for a groundwater sample containing 37.7 µg As/L.

Table 6 shows characteristics of dynamic adsorption studies using industrial waste and by-products as adsorbents.

Table 6. Comparison of dynamic tests using industrial waste and by-products as adsorbents for arsenic removal.

Adsorbent	Treatm.	As Species	Influent Concentration (µg/L)	Max. Adsorption Capacity (mg/g)	As Remov. (%)	Breakth. Time at 10 µg/L (h)	BV to Breakth. Point of 10 µg/L	Ref.
Bagasse fly ash (BFA)	None	As(V)	50		98.9			[107]
		As(III)	50		95.6			[107]
Water treatment residual solids	None	As	37.7				26,400	[108]
Iron oxide-coated cement (IOCC)	$Fe(NO_3)_3{\cdot}9H_2O$	As(V)	2000–4000	3.8852		4.0–41.5		[109]
		As(III)	500–2700	0.1462–0.3082	54.04–84.77			[110]
Hardened paste of Portland cement (HPPC)	Cement + water	As(V)	500		>90			[111]
Stainless steel slags	None	As(V)	10,000		84%			[112]

IOCC presented an adsorption capacity similar to other traditional adsorbents such as carbon-based materials and metal (hydr)oxides. In terms of arsenic removal efficiency, the iron modified cement (IOCC) does not present advantages compared to the hardened paste of Portland cement (HPPC), even the studies being conducted with different arsenic species. Use of industrial waste and by-products can be an economic and sustainable way to remove arsenic from water, but further studies should be carried out on their viability for practical applications.

3.8. Nanocomposites

Nano adsorbents are very promising and highly efficient materials for arsenic removal from groundwater; however, separation and regeneration are still challenges to their practical application [11]. Nanoparticles are <100 nm in size [113], and different nanomaterials have been tested for arsenic adsorption. Column studies have been conducted with nanoparticles of different materials: alginate [101], aluminum [114], carbon [115–117], copper [118,119], iron [21,96,120–122], manganese [123], polymers [114,124,125], zirconium [126], and titanium [21,127,128]. In order to facilitate reuse and regeneration, the nanoparticles are usually coated onto another materials such as glass beads [127], gel beads [114,125], polystyrene [124], and alginate beads [82].

Concrete-based iron oxide (IO) nanoparticles (NP) were tested in laboratory experiments using a glass column of 2.2 cm in diameter and different bed heights (10, 20, and 30 cm) [122]. Contaminated water containing 10 mg/L of As(V) was fed constantly at 7.5 mL/min flow rate. The time of breakthrough for 10 µg/L of arsenate was found to be 4.1, 6.5, and 8.6 h for 10, 20, and 30 cm of bed height, respectively.

Multi-walled carbon nanotubes (CNTs) were also investigated in column adsorption tests for As(III) and As(V) removal [115]. A glass column containing 2.0 g of CNT and 2.0 g of washed river sand was operated under different flow rates (20, 30, and 40 mL/min) at pH 6, with an initial arsenic concentration of 40 µg/L. The optimal flow rate for operation was found to be 30 mL/min leading to an adsorption capacity of 0.014 mg/g for As(V) and 0.0135 mg/g for As(III). The regeneration process was conducted with different eluents, and 1.0 M HCl achieved the best results.

A more recent study was conducted with another form of carbon-based sorbent: multi-functionalized magnetic graphene [117]. Adsorption-desorption column tests for As(III) and As(V) were taken with different flow rates, using NaOH and HCl as eluents, and results showed an arsenic recovery of approximately 98%.

Table 7 presents the results of dynamic adsorption studies conducted with nanocomposites. They are claimed to be a highly efficient option for metals and metalloids adsorption; however, the results of tests developed in continuous mode do not prove that yet.

Table 7. Comparison of dynamic tests using nanocomposites, GFH, and commercial adsorbents for arsenic removal.

Adsorbent	Arsenic Species	Influent Concentration (μg/L)	Max. Adsorption Capacity (mg/g)	Breakthrough Time at 10 μg/L (h)	BV to Breakthrough Point of 10 μg/L	Ref.
			Nanocomposites			
Microwave-assisted economic multi-walled carbon nanotubes (MWCNTs)	As(V) As(III)	40	0.014 0.0135	50–100		[115]
Iron doped phenolic resin-based activated carbon micro-nano particles	As(V) As(III)	1000 1000–3000	2.78–3.24 2.60–3.30			[116]
Cupric oxide (CuO) nanoparticles	As	109		7–15		[119]
α-MnO$_2$ nanofibers	As(V) As(III)	200			120 200	[123]
Concrete-maghemite nanocomposites	As(V)	10,000		4.1–8.6		[122]
Metal-doped titania nanoparticles coated glass beads	As	250–1000	0.477–0.61	1.67–6.5		[127]
	As(III)	250		1–3.67		[127]
Zirconium-organic frameworks@biomass-derived porous graphitic nanocomposites (Ui)	As(III) As(V)	500 500		1.667 2.167	500 651	[126]
			Granular ferric hydroxide (GFH) and commercial adsorbents			
GFH	As(V)	800	4.92–6.07			[129]
	As	10,000			~500	[130]
	As	500			More than 40,000	[130]
Ferrosorp plus (FP)			0.79		728 [1]	[131]
ArsenX^{np}—a hybrid anion exchanger	As	85			29,000 [2]	[120]
Adsorbsia GTO	As	28			3000–10,000	[128]
Iron nanoparticle resin (Lewatit FO36)	As(V)	500	3.229		3512–4000	[132]
Fe (III)-phosphorylated resin (Fe-XAD8-DEHPA	As	80		6 (for 3 μg/L)		[133]

[1] For breakthrough point of 3.75 μg/L [2] For breakthrough point of 50 μg/L.

3.9. GFH and Commercial Adsorbents

Granular ferric hydroxide (GFH) is a well-known, commercially available adsorbent used for arsenic and other water pollutants removal; however, it has some limitations for long-term application [134]. Column studies have been conducted to investigate specific characteristics of an arsenic adsorption process with GFH [13,129–131,135–140].

Rapid small-scale tests were performed using three commercially available adsorbents—E33, GFH, and Metsorb—to assess the interference of silica, phosphate, vanadate, and pH in each of them [137]. The GFH was found to be the most susceptible to ionic interference. In addition to that, the pH and silica had the greatest impact on arsenic adsorption compared to vanadium and phosphate. Another lab-scale study showed that organic matter reduced the adsorption capacity of GFH [129]. Real-time pilot-scale tests were conducted with GFH treating an inlet arsenic concentration of approximately 1000 µg/L at a flow rate of 25 mL/min, and X-ray fluorescence (XRF) was validated as a monitoring method for arsenic accumulation through the column [136].

Resins are also popular commercial adsorbents due to their excellent hydraulic properties and mechanic resistance [133]. The combinations of resins with metal (hydr)oxides are usually known as hybrid systems. They gather the selectiveness of the metal with the characteristics of the resins to result in more efficient products with better adsorption capacity.

A commercial hybrid anion exchanger named ArsenXnp is formed by the combination of anion exchange resin beads with hydrated ferric oxide (HFO) nanoparticles [120]. Field columns packed with ArsenXnp could run more than 20,000 bed volumes before the breakthrough point of 50 µg/L of arsenic. The regeneration process after saturation was carried out using 2% NaCl and 2% NaOH and allowed multiple cycles of reuse.

Table 7 shows characteristics of dynamic adsorption studies conducted with GFH and commercial adsorbents. Particularly good results were achieved in terms of treated bed volumes, using some alternative materials, such as biosorbents and industrial waste. They can treat similar amounts of contaminated water, in a more sustainable and economic way.

3.10. Layered Double Hydroxides (LDH)

Layered double hydroxides (LDH) are a class of anionic clays that have been employed more recently to remove oxyanions such as As(V) and As(III) from aqueous solution [141]. Despite their good adsorption characteristics, the use of LDH in fine powder and its further separation after treatment is still a hindrance for their wide application [142]. There are various forms of LDH; however, few of them have been applied in dynamic tests for arsenic removal.

A zirconium and iron layered double hydroxide (Zn-Fe-LDH) was prepared by a modified co-precipitation method, and its arsenic removal capacity was studied in column tests [143]. A column (2.0 cm diameter × 10 cm height) packed with 0.8 g of adsorbent was fed with As(V) solution of 1 mg/L, at a flow rate of 0.6 mL/min and pH 7. The handling capacity achieved for the breakthrough point of 10 µg/L was 12.5 L/g of adsorbent. Desorption tests showed that a solution of 0.5 M NaOH was able to remove 67.8% of As(V) from the adsorbent.

Fixed-bed experiments were also conducted with calcinated Mg-Al-LDH to investigate the effect of particle size, pH, initial As(V) concentration, and flow rate in breakthrough curves [144]. The decrease in particle size, flow rate, and initial concentration all caused an increase in breakthrough time.

Results of adsorption studies, conducted with LDH adsorbents in continuous mode, are presented in Table 8. Although LDH are composed of metals with strong affinity to arsenic, the reported column studies did not show interesting results if compared with other metal-containing adsorbents discussed in this article.

Table 8. Comparison of dynamic tests using LDH as adsorbents for arsenic removal.

Adsorbent	Arsenic Species	Influent Concentration (µg/L)	Breakthrough Time at 10 µg/L (h)	BV to Breakthrough Point of 10 µg/L	Ref.
Zn-Fe-LDH	As(V)	1000		~300	[143]
Mg-Al-Cl-LDH	Total As	506	10		[142]
Mg-Fe-Cl-LDH	Total As	506	7		[142]

4. Recovery and Regeneration

The recovery of arsenic and regeneration of the adsorbent are relevant to ensure the viability of the adsorption process on a large scale. The balance between the cost of regeneration and the cost of new adsorbent should be taken into consideration when choosing adsorption for arsenic removal from contaminated waters. Some of the studies discussed in the previous sections presented the results of adsorption/desorption cycles in continuous mode to evaluate the increase in adsorbent life span.

Most desorption experiments have been conducted using NaOH [7,20,33,42,47,50–52,60,70,74,76,78,81,90,93,96,100,117,127,132,140] as eluent with different concentrations, showing that arsenic recovery is possible with a basic solution. However, some studies reported that alkaline treatment removed coated iron from the adsorbent, and considering that most treatments are iron-based, the concentration of eluent should be adjusted depending on each adsorbent's characteristics. Other chemicals were also tested in the recovery process: sodium bicarbonate ($NaHCO_3$) [88], sodium fluoride (NaF) [80], sodium chloride (NaCl) [71,80], potassium hydroxide (KOH) [95], monopotassium phosphate (KH_2PO_4) [41], phosphoric acid (H_3PO_4) [72,73], hydrochloric acid (HCl) [72,73,85,117], and sulfuric acid (H_2SO_4) [72,73].

Some adsorbents were successfully regenerated after 320 cycles [73], and others after only two cycles [75,76]. Good arsenic recovery rates were observed in column studies. For example, with soils and rocks, recovery ranged from 92% to 99% [51,52,55,59]. Some biosorbents and nanocomposites also presented higher arsenic recovery, such as rice polish (99.6%) [93] and cupric oxide nanoparticles (99.4%) [119].

5. Disposal of Contaminated Adsorbents

The sustainable application of novel adsorbents on a real scale also relies upon the fate of the contaminated bed. The toxicity characteristic leaching procedure (TCLP) is commonly applied to evaluate whether the waste can be safely disposed. According to USEPA 1992, the disposal of arsenic waste can occur in domestic landfills if the leachate concentration of arsenic is less than 5.0 mg/L; otherwise, the waste should be disposed of in a hazardous waste landfill [74]. TCLP results reported with arsenic-contaminated beds are presented in Table 9.

Table 9. TCLP results from arsenic-contaminated beds.

Type of Adsorbent	Adsorbent	Maximum Arsenic Leached (mg/L)	Under USEPA Limit?	Ref.
Biosorbent	Ferric hydroxide microcapsule-loaded alginate beads (FHMCA)	0.3	Yes	[82]
Biosorbent	Iron doped chitosan spacer granules (ICS)	0.02	Yes	[76]
Biosorbent	Magnetic binary oxide particles (MBOP) (solidified and stabilized)	0.07	Yes	[74]
Carbon-based	Iron-containing granular activated carbon	0.09	Yes	[67]
Carbon-based	HFO-PF-coated GAC	<0.1	Yes	[66]
Commercial	ArsenX^{np}—a hybrid anion exchanger	<1.0	Yes	[120]
Metal (hydr)oxide	Iron mineral-quartz sand	0.4	Yes	[16]
Mineral	Natural siderite	0.4	Yes	[45]
Mineral	Granular schwertmannite	0.025	Yes	[46]
Mineral	Activated siderite-hematite	0.3	Yes	[39]

In addition to waste landfill disposal, studies reported other destination alternatives: the incorporation of arsenic sludge in construction material, the mixing of arsenic waste from biomass origin with coal for energy generation, and the mixing with livestock waste to be converted by microorganisms into gaseous arsine [145].

6. Conclusions

Much research has been conducted in the past decades to develop adsorption systems for continuous arsenic removal from contaminated water. Different types of materials have been tested and improved to perform better adsorption. Maximum adsorption capacities of 28.96 mg/g and 74.8 mg/g were reported mostly by novel adsorbents like binary metal oxides and biosorbents, respectively. Most studies were preliminary, and there is still a gap between the identification of novel adsorbents and feasibility studies for practical applications.

The other materials presented similar values of maximum adsorption capacities: 6.07 mg/g for GFH, 5.00 mg/g for minerals (mill-scale derived magnetite particles), 3.8852 mg/g for industrial waste (iron oxide-coated cement), 3.51 mg/g for soil and rock (pisolite), 3.30 mg/g for nanocomposite (iron doped phenolic resin/AC), 3.229 mg/g for commercial (Lewatit FO36), and 2.873 mg/g for carbon-based (Fe/GAC),

Biosorbents were employed in most studies published, followed by metal oxides and nanocomposites. There was an increase in the last years in research for low-cost adsorbents such as biosorbents and industrial waste and by-products, which is of high importance to fill the gap in efficient and cheap treatment in most arsenic-affected places.

Arsenic adsorption in continuous mode is mostly affected by pH, temperature, flow rate and hydraulic retention time, adsorbent quantity (bed height), adsorbate initial concentration, and the presence of other chemical elements. Iron incorporation is the most common treatment to increase the arsenic adsorption capacity of adsorbent materials, followed by other metals such as aluminum and zirconium.

Desorption, like adsorption, is dependent on parameters such as eluent type and concentration, pH, flow rate, and retention time. For each arsenic adsorption system, those parameters should be adjusted to result in better regeneration of adsorbent and recovery of adsorbate.

Arsenic-contaminated adsorbents can be non-hazardous, and a variety of disposal alternatives are being investigated for the fate of arsenic waste. However, the topic should be the focus of deeper studies, especially in the testing of the viability of exhausted adsorbents alternative applications on a real scale, including more research into the toxicity and stability of materials incorporating arsenic adsorbent waste.

Further adsorption investigation must be conducted at pilot-scale, which is important to evaluate the application of the adsorbent in full scale, including more studies in regeneration and recovery, which enhance the economical balance of adsorption. Furthermore, the calculation of treatment costs and life cycle assessment (LCA) of the adsorption systems are relevant to be compared with other water treatment techniques.

Overall, this work gathered the most relevant worldwide research into low-cost and alternative arsenic adsorption dynamic systems, which is important to help to consolidate the available knowledge for sustainable solutions that provide safe access to potable water in contaminated areas.

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