Review

Arsenic Removal from Contaminated Water Using Natural Adsorbents: A Review

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Abstract: More than 170 million individuals have been influenced by arsenic (As) because of the ingestion of As-polluted groundwater. The presence of As in water bodies, particularly groundwater, has been found to become a widespread issue in the past few decades. Because arsenic causes extreme wellbeing impacts, even at a low concentration in drinking water, the innovations of As removal from contaminated water are of significant importance. Traditional strategies, for example, reverse osmosis, ion exchange, and electro-dialysis are generally utilized for the remediation of As-polluted water; however, the high cost and/or sludge production restricts their application in less-developed areas. The utilization of adsorbents acquired from natural materials has been explored as an alternative for the costly techniques for As removal. This paper aims to review the past and current developments in using natural adsorbents or modified natural materials for arsenic removal and show the different parameters, which may influence the As removal effectiveness of the natural adsorbent, such as contact time, adsorbent dosage, flow rate, pH, reusability, temperature, and influence of others ions.

Keywords: adsorption; arsenic; natural adsorbents

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1. Introduction

The name arsenic (As) is thought to originate from the Syriac, Arabic, or Persian languages, and was later adopted into the Greek word arsenikon. The yellow-colored orpiment (As₂S₃) is infamous as the lord of all poisons. Arsenic (As) is an exceptionally harmful and cancer-causing metalloid that accounts for approximately 0.00005% of the Earth's outside layer. As ranks as the twentieth among the elements in abundance in the Earth's crust, fourteenth in the seawater, and twelfth in human body [1]. Arsenic occurs in rock soils, organisms, natural water, air, and in the other natural environments [2]. The element of As can form over 200 minerals in soil, and mainly exists as As(0), As(III), As(V), and arsenide. Besides, a high As concentration is found in numerous oxide minerals and hydrous metal oxides, either as a part of the mineral structure or as adsorbed species. The amount of As in sedimentary rocks ordinarily ranges from 5 to 10 mg/kg, which is usually over the normal content in a terrestrial environment, approximately 1.5 to 3 mg/kg [3]. A significant amount of As in natural water is also from anthropogenic sources, such as cosmetics, mining operations, coal combustion, smelting, pesticides, dyes, wood treatment, As trioxide stockpiles and paints. Arsenic is also highly soluble in water [4–6].

The two most prevailing types of As in aqueous environment are As(III) and As(V). Depending on the environment conditions, such as pH, the redox condition, the microbial activity, and presence of complexing ions, the pentavalent As mainly consists of arsenate ions (H₂AsO₄^{1−}, HAsO₄^{2−}, AsO₄^{3−}) and arsenic acid (H₃AsO₄). Similarly, the As(III) species may present as arsenite ions (H₂AsO₃^{1−}, HAsO₃^{2−}, and AsO₃^{3−}) and arsenious acid (H₃AsO₃). The pentavalent species are the predominant species under aerobic con-

ditions, such as surface water [7], whereas the trivalent species are the prevailing species under anoxic conditions, such as subsurface groundwater [8,9].

2. Harmful Effects of Arsenic

As(III) and As(V) are both very toxic, with increased risk for cancer of the skin, lungs, urinary bladder, liver, and kidney [10], but As(III) is multiple times more harmful and versatile than As(V). Wellbeing impacts of arsenic rely upon different kind of parameters. These include the type and amount of arsenic that has been swallowed, how long you have been exposed to arsenic, and how the body reacts to arsenic. Unborn infants, children, and people with long sicknesses and the elderly are the weakest to arsenic exposure. The impacts of arsenic, when ingested even in limited quantities, show up gradually; it might take quite a long while for the harming effects to become obvious.

Arsenic pollution in natural water is an overall issue, and has become a significant issue and challenge for world specialists, researchers, and even politicians. For instance, persistent arsenic poisoning due to drinking arsenic-polluted water has been one of the terrible wellbeing impacts influencing eight areas of West Bengal since the mid 1980s. Itemized clinical assessment and investigation of 248 such patients showed the clinical signs of such harmfulness. Well beyond hyperpigmentation and keratosis, powerlessness, paleness, painful eyes, leg swelling, liver fibrosis, persistent lung ailment, gangrene of toes, neuropathy, and skin malignant growth were a portion of different symptoms [11]. The World Health Organization (WHO) and the U.S. Environmental Protection Agency (US EPA) have limited the As concentration level at 10 µg/L in 2001.

3. Arsenic Removal Technology

Considering the danger of As in water, especially in drinking water, many approaches have been studied for As removal. Traditionally, there are a few techniques including membrane filtration, coagulation and flocculation, electrochemical techniques, bioremediation, ion or particle exchange, and adsorption utilized in arsenic removal [12–15].

3.1. Membrane Filtration

Membrane filtration is most commonly used to eliminate arsenic from water. Membrane films are regularly manufactured materials with billions of pores or minute openings that operate as a specific obstruction; the structure of membrane permits a few constituents to go through, while others are avoided or dismissed. The different kinds of membrane filtration can be divided into four techniques: ultrafiltration membranes are usually with 0.0003 to 0.1 microns for colloids, viruses, and certain proteins removal [16]; nanofiltration with 0.001 to 0.003 microns for physical removal based on both charge and molecular size; microfiltration with 0.1 micron for bacteria and suspended solids removal [17]; and hyperfiltration or reverse osmosis with 0.0005 microns for desalinization [18].

3.2. Coagulation

Many studies have showed that coagulation can be used for removing arsenic from contaminated water by the addition of a coagulant in the arsenic solution. This process can also remove many other suspended and dissolved constituents from water, such as fluoride, manganese, iron, phosphate, and turbidity. This arsenic removal technique is profoundly reliant upon pH, the valence of the arsenic species, dosage of coagulant, and the initial arsenic concentration [19].

3.3. Electrochemical Techniques

The utilization of electricity for waste water treatment started early, since 1887 in the United Kingdom [20]. There are a lot of different electrochemical techniques, including electrodeposition [21], electrocoagulation [22,23], electrodialysis [24], electroflotation [25], and electrooxidation [26] used for heavy metal removal. Electrocoagulation is a process using chemical and physical reactions in which sacrificial electrodes are used for the

generation of coagulants. This process, needing a high amount of electricity, is the one used the most for arsenic removal from water.

3.4. Bioremediation

Some natural organisms, such as bacteria, algae, and plants especially the aquatic plants, have a capacity to amass trace elements (heavy metals), which allows them to be used for the treatment of polluted water from industrial or agrochemical discharges, especially contaminated with heavy metals [27,28].

3.5. Ion or Particle Exchange

Ion exchange is a physical/chemical process in which particles held electrostatically on the outside of a solid phase are exchanged for particles of same charge in a solution (i.e., drinking water). The solid is ordinarily a synthetic or natural anion exchange resin, which is utilized to specially remove specific contaminants of concern. Ion exchange is generally utilized in drinking water treatment for softening (i.e., removal of magnesium, calcium, and other cations in exchange of sodium), as well as removing arsenate, nitrate, selenate, and chromate from municipal water [29].

3.6. Adsorption

Adsorption is a process that utilizes solids for eliminating substances from either liquid or gaseous solutions, by attraction on the solid's surface. Solids, such as synthetic materials (fibers, resins, activated carbon, metal hydrides, membranes) [30,31] or natural materials [32] are widely utilized in industrial application for wastewater treatment and purification. However, synthetic materials are expensive to be produced, as the production of them requires a lot of energy and is not environment friendly compared to natural materials.

As mentioned above, most processes have been proved to be effective in removing As from water. For example, As in water can be effectively removed after being blended with other metal oxides (iron or aluminum oxides) for a coagulation to change into the solid phase [33]. However, the high energy required (electrocoagulation), the high cost of their process (membrane filtration), and the non-environmentally friendly residue (coagulation and synthetic adsorbent adsorption) after the treatment processes, limit their application and lead researchers to conduct new research with natural materials, which are ecological and cheaper to find [34–36]. In this paper, we reviewed the removal performances of As from contaminated drinking water/groundwater by adsorption using natural adsorbents and/or modified natural materials.

4. Biosorbents

Biosorbents (natural adsorbents) are made from natural materials, including natural fibers, volcanic rocks, soils, plant biomass, agricultural and industrial wastes, animal shells, microalgae, and fungal biomass (Figure 1). A common feature of these materials is that their large specific surface area, which is capable of physically retaining arsenic ions or molecules. In recent years, more and more natural adsorbents are used because of their higher arsenic removal capacity, reusability, and lower cost and environment impacts as compared with the other methods involving synthetic membranes and materials using significant chemical dosage [37,38]. Among all the natural materials, agricultural wastes, comprised of proteins, extractives, hemicellulose, lignin, lipids, starch, and simple sugars, contain more functional groups, which facilitate heavy metal retention [39–42]. In Table 1, some different natural adsorbents that have been used for As removal from water are listed.

Figure 1. Different types of natural adsorbent As.

Table 1. Some As natural adsorbents and their adsorption capacities.

Adsorbents	Treatments	Isotherm Model Fit	pH	Adsorption Capacities mg/g	
				As(III)	As(V)
Wheat Straw [43]	NaHCO ₃	Langmuir	7	No data	0.097
Wheat Straw [44]	FeSO ₄ and FeCl ₃	Langmuir	6	No data	30.24
Black Tea [45]	No Treatments	Freundlich and Langmuir	6	No data	1.76
G. cambogia fruits rinds [46]	IICT Technology	Langmuir	6	128.10	No data
Coconut Fibers [47]	HNO ₃ and NaOH	Freundlich	4	0.12	No data
Sugarcane bagasse [48]	(Fe(NO ₃) ₃ ·9H ₂ O)	Langmuir	6	0.6	No data
Sawdust [49]	Fe(III)	Langmuir	8	No data	5.8
Orange Peel [50]	No Treatments	Freundlich	7	No data	4.8
Moringa Lamarck seed [51]	No Treatments	Langmuir	8	1.5	2.1
Rice Husk [52]	No Treatments	Freundlich	7	220.1	No data
Mango leaf powder [52]	No Treatments	Freundlich	7	250.07	No data
Mosambi Citrus Peel [53]	No Treatments	Freundlich	6	2.12	3.32
Rice straw [54]	Fe(NO ₃) ₃	Langmuir	4	No data	21.739
Rice husk [55]	Fe(III)	Langmuir	4	No data	2.47
Corn cob husk [56]	FeCl ₃	Langmuir	6	50	No data

Table 1. Cont.

Adsorbents	Treatments	Isotherm Model Fit	pH		Adsorption Capacities mg/g	
					As(III)	As(V)
Coconut coir pith (CP) [57]	Epichlorohydrin and dimethylamine	Langmuir	7		No data	13.57
Groundnut shells [58]	No Treatments	Langmuir	8		0.014	No data
Bead Cellulose (Cotton) [59]	(FeCl ₃ ·6H ₂ O)	Langmuir	9	4	99.6	33.2
Sawdust [60]	ZrOCl ₂ ·8H ₂ O	Langmuir	9	4	29	12
Rice husk [61]	Iron oxide	Langmuir	6		No data	82
Paddy Husk Ash [62]	AlCl ₃ ·6H ₂ O	Freundlich	4		No data	0.063
Zeolite stones [63]	Al ₂ (SO ₄) ₃ ·16H ₂ O	Freundlich	6		No data	208
Iron oxide-coated sand [64]	Fe(III)	Langmuir	7.5		28.57	No data
Goethite [65]	No treatment	No data	5		No data	1
Iron-coated zeolite [66]	FeCl ₃	Langmuir	4		No data	0.68
Iron-modified activated carbon [67]	Fe(NO ₃) ₃ ·9H ₂ O	Langmuir	8	6	43.6	51.3

5. Treatment of the Natural Adsorbents

Some natural adsorbents without any kind of pre-treatment are good enough for removing As from water because of their surface structure composition, such as the acidic functional groups (–SH, –COOH) present in the rice husk cellulose component or the Fe and Al oxide present on montmorillonite, kaolinite, and illite clay minerals as impurities (Figure 2). These functional groups may facilitate the interaction with As anions [68,69]. On the other hand, as we can see in the Table 1, some adsorbents need to be pre-treated to improve their As adsorption capacity. The goal of pre-treatment by washing and cleaning, using distilled water, alkali (NaOH) or acidic (HCl) solution is to remove impurity particles or add some functional group on the surface of the natural material. Therefore, pre-treatment will make it possible to highlight the functional groups (CH₃–OH, –COOH, CH₃–NH₂, Fe(OH)₃, Al(OH)₃), and therefore will facilitated the interactivity between natural adsorbents and As ions.

Distilled water is used most of the time to remove surface adhered impurity particles, water-soluble materials [57]. The distilled water washed materials will be dried under specific conditions as necessary, for example, coconut coir pith (CP) dried at 80 °C, groundnut shell dried under sun for 2 days [58], and rice husk dried at 60 °C [70]. Alkali pre-treatment with 20% NaOH solution is used to degrease cotton and add hydroxyl group onto the surface [59], and remove the lignin of sawdust [60]. Acidic pre-treatment selectively removes the organic base by converting it into a water-soluble salt as HCL (10%), which is used to clean rice husk to remove all the impurities, then later crushed and dried at 500 °C in muffle furnace for 8 h [61], and *Dialium guineense* seed shells have been crushed then soaked in (40%) phosphoric and nitric acid, then heated to a carbon activation temperature of 400 °C for 30 min.

Figure 2. Functional groups existing on the surface of natural adsorbents.

The objective of the treatment is to change the surface structure, to improve the As removal capacity of natural adsorbents by adding essential functional groups (Fe^{3+} , Al^{3+} , TiO_2) [71–74]. Aluminum ions are often used for the modification of the natural adsorbent's surface, since Al^{3+} oxidative capacity towards As(III) is well known [75,76]. The reactions below might be the interaction of As(V) and As(III) with the natural adsorbent (M) after its surface modification with aluminum:

As(V)

As(III)

For example, hydroxyl-alumina has been used as a surface activating agent for the modification of paddy husk ash particles, using $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ and aluminum fine powder [62]. The surface of crushed pumice and zeolite stone particles can be coated with aluminum ions by immersing pumice and zeolite stone particles in 0.5 M $\text{Al}_2(\text{SO}_4) \cdot 16\text{H}_2\text{O}$ solution, and drying at room temperature for 72 h [63]. Similarly, the removal of As(V) and As(III) can be enhanced with the introduction of ferric ions on the surface of natural materials (M) because of the formation of ferric arsenate $\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$ at a low pH [67,77,78]. The reactions below might be the interaction of As(V) and As(III) with the natural adsorbent (M) after its surface modification with iron:

As(V)

As(III)

Maji et al. [79] and Joshi et al. [80] found that the surface structure of crushed natural rock particles and river sand could be successfully modified using Fe(III) nitrate solution,

then dried in 110–150 °C hot air; this finding was similar to Jeon C.S. et al. [66] using zeolite (clinoptilolite) as a natural material. In some cases, natural iron oxide minerals, such as magnetite (FeOFe_2O_3), hematite (Fe_2O_3), goethite ($\text{FeO}(\text{OH})$), and laterite, were used directly for As adsorption from water without any pre-treatment or treatment [65].

6. As Removal Performance

Many researches have been conducted to evaluate the As adsorption performance of many different types of natural adsorbents, by studying the effects of different factors [81]. There are many parameters that can influence the removal capacity of the natural adsorbents, such as pH, initial dosage of the adsorbents, initial concentration of the As solution, contact time, temperature, and the effect of other ions in the water solution. The natural adsorbent for As removal has mainly been tested by using two different methods, batch test and column test [82–86].

6.1. Overall Adsorption Performance

To achieve an efficient As removal from water, both the initial As concentration and adsorbent dosage are optimized before conduction water treatment [87,88]. As can be seen in the Table 2, it is obvious that an increase in the initial As concentration leads to a decrease in the As removal efficiency [89,90]. Usually, As removal efficiency increases with the increase in the adsorbent's dosage, due to the higher surface area or exchangeable sites provided. However, if the adsorbent's dosage is too high, the removal efficiency of As will finally stay constant, which is not economic [91–94]. Take iron-modified peat for example, it was determined that only 60% of the As(V) was removed with an initial concentration of $800 \text{ mg} \cdot \text{L}^{-1}$, and the ratio reached more than 90% when its initial concentration decreased to $300 \text{ mg} \cdot \text{L}^{-1}$ [95,96]. Compared with iron-modified peat, PAC-500 (peels activated carbon) and PPAC-500 (pulp activated carbon) seem to have a lower As adsorption capacity. It was determined that when the initial concentration of As was around 2.5 mg/L , less than 80% of As was removed, and this ratio would be above 95% when the initial concentration of As decreased to around 0.5 mg/L [97]. Again, considering only 40–50% of As was adsorbed when its initial concentration was in the range of $0.005\text{--}0.02 \text{ mg/L}$, modified clinoptilolite zeolite (MCZ) seems have the lowest As adsorption capacity [98].

Table 2. Some studies related to initial As concentration and adsorbent dosage impact on As adsorption capacity.

Adsorbent	Adsorbent Dosage (g)	Solution Quantity (mL)	Initial Concentration (mg/L)	Removal (%)	
				As(V)	As(III)
Peat-based sorbents [96]	0.5	40	8–904	No data	100–43
Red mud-modified biochar (RM-BC) [87]	0.12	30	1–50	100–32	50–4
Novel magnetic chitosan nanoparticle (MCNP) [89]	0.05	100	0.2–50	100–65	100–60
Hematite Pinewood biochar (HPB) [99]	0.05	20	1–50	25–2	No data
Modified clinoptilolite zeolite (MCZ) [98]	1	200	0.005–0.05	55–40	No data
Iron-modified peat [96]	0.5	40	100–270	100–70	No data
Biomass of <i>Citrus limmeta</i> (PPAC-500) [97]	0.15	50	0.05–2.5	100–62	93–55.2
Leaves of <i>P. roxburghii</i> powder [88]	0.2–2	50	10	4–75	No data
Iron modified montmorillonites [100]	0.025–0.3	25	0.005	97–100	96–100
<i>Corynebacterium glutamicum</i> MTCC 2745 biofilm supported on Neem leaves NL/ MnFe_2O_4 composite [93]	0.01–0.1	100	50	79–82.5	72–77
<i>Bacillus arsenicus</i> biofilms supported on a Neem leaves/ MnFe_2O_4 composite [94]	0.01–0.1	100	50	86–89	79–83
<i>M. charantia</i> plants biomass [91]	0.05–0.25	50	0.5	No data	66–88
Bone char [101]	0.05–0.4	500	0.5	30–100	No data

6.2. Effects of Contact Time on As Adsorption

The evaluation of operational parameters, such as the contact time (batch adsorption) and flow rate (column adsorption) are also important to determine the time at which the adsorption reaches equilibrium. The quantity of As adsorbed per unit mass of adsorbent, and the percentage of As adsorbed will be determined respectively using Equations (13) and (14) given below:

$$q_t = ((C_0 - C_t)/M) \times V \quad (13)$$

$$A(\%) = ((C_0 - C_t)/C_0) \times 100 \quad (14)$$

where, q_t is the quantity of As adsorbed at time t ; $A(\%)$ is the percentage of As adsorbed at time t ; C_0 is the initial As concentration; C_t is the concentration of As in the aqueous phase at time t ; V is the volume of the aqueous solution; and M is the mass of natural As that will be used in the experiment [102]. The increase in the flow rate causes a decrease in the contact time between the adsorbent and As solution, which in turn lowers its removal efficiency. If the contact time is not long enough, the surface of the adsorbent will not be able to be charged enough, which will be a waste of the adsorbent. On the other side, if the contact time is longer than the equilibrium time, the adsorbent might start releasing back some As particles in the aqueous solution, which will not benefit the removal efficiency [103,104]. Frequently, the quantity of As adsorbed increases rapidly because there are still many adsorption sites on the natural adsorbent surface at first, then it will gradually slow down until it stays constant after the equilibrium adsorption time [105–109].

The augmentation of flow rate on As adsorption by thioglycolated sugarcane carbon (TSCC) and iron-modified cement slurry have been studied. The collected data showed that the As removal performance decreased with the increase in the flow rate from 3.0 to 7.0 mL min^{−1} and 5 to 14 mL/min [103,110]. However, the results using (MNCF) modified nonwoven cotton fabric, modified rice husk particles for As removal from water, showed that within 1 h a high As adsorption rate (almost 80%) was noticed [55,111]. The marine sand might have fast As adsorption in comparison to algae, as zirconium oxide-coated marine sand (ZrOCMS) removed more than 90% within 75 min, which is quick [112] compared to bio-adsorbents, such as algae (*Lessonia nigrescens*), in which the maximum adsorption capacity was reached after 300 min [113].

6.3. pH

The adsorption of As ions species are strongly governed by the pH of the aqueous solution. The pH of the solution is an important parameter, which controls the As species present in water and the natural adsorbent surface chemical composition [114]. The distributing equation of As(III) is as follows:

The distributing equation of As(V) is as follows:

Mostly, if the As removal is through chemical adsorption, the As(III) adsorption is better at a high pH [115] because at pH 1–7 the predominant As(III) species H_3AsO_3 ($\text{pK}_a = 9.2$) is uncharged, which can negatively impact the As removal performance. As shown in Figure 3, the adsorption performance of As(III) increased notably with the increase in the solution pH, and the best solution pH for As(III) adsorption was around

pH = 8.5 when the copper impregnated coconut husk carbon (CICHC) was used as the adsorbent [116]. A study using fly ash from the power plant after burning biomass and coal for As(III) removal showed that the best removal data was around pH 12 [117]. However, for As(V), its removal is mostly better at a low pH because the adsorption performance decreases with the diminution of H_2AsO_4^- ($\text{pK}_a = 7$) percentage in the aqueous solution [81,118–122]. As it can be seen in the Figure 4, Wang et al. [123], using Ni/Fe modified loblolly pine (*Pinus taeda*) wood biochar (NFMB) for As(V) removal, showed that the adsorption capacity decreased with the solution pH rising from 3 to 9. Another study using iron-modified loblolly pine (*Pinus taeda*) wood biochar (nZVI/BC) for As(V) removal showed that the removal efficiency decreased briskly at pHs 3–3.7, and kept dropping as the solution pH increased [124].

6.4. Reusability

Desorption and regeneration are important parameters because a good natural adsorbent is an adsorbent that can remove enough As from water and can be reused without losing its adsorption performance too much, which indicates that the adsorbent can be recycled and reused. The loss of the adsorption capacity, after much desorption and regeneration, is due to the loss of adsorption sites on the adsorbent surface [90,123,125,126]. Generally, it can be noticed in the Table 3, most of the natural adsorbents have good reuse performance. It was determined that after the iron hydroxide/manganese dioxide doped straw activated carbon (Fe-Mn-Sac) was used for three adsorption–desorption cycles, the adsorption capacity of Fe-Mn-Sac had a negligible variation (from 85% to 78%) [127]. For iron-modified water hyacinth biochar, the arsenate removal percentage decreased from 100 to 65% after four cycles of regeneration, which was not a bad performance regarding the high arsenate initial concentration in the water (5 mg/L) [128].

Figure 3. As(III) adsorption as a function of pH variation [116,117].

Figure 4. As(V) adsorption as a function of pH variation [123,124].

Table 3. Studies on some natural adsorbent's reusability performance.

Adsorbent	Adsorbent Dosage (g)	Initial Concentration (mg/L)		Regeneration	Adsorption Capacity (%)		Reference
		As(V)	As(III)		As(V)	As(III)	
The green alga (<i>U. cylindricum</i>) biomass [129]	0.1	10		10	No data	96–93	Tuzen M. et al., 2009
Ni/Mn-layered double hydroxide (LDH) biochar (NMMB) [123]	0.1	40		3	100–98	No data	Wang S. S. et al., 2016a
Ni/Fe layered double hydroxide (LDH)-biochar (NFMB) [130]	0.1	50		3	100–92	No data	Wang S. S. et al., 2016b
Charred orange peel (COP) [131]	0.2	200		3	100–90	No data	Abid M. et al., 2016
Fe-Mn-straw biochars [127]	1	20		3	No data	85–78	Xiong Y. et al., 2017
Xanthated water melon rind (X-WMR) [132]	0.2	As(V) 5	As(III) 4	4	100–50	100–20	Shakoor M. B. et al., 2018
Magnetite-modified water hyacinth Biochar (MW250 ₁) [128]	0.2	5		4	100–50	No data	Zhang F. et al., 2016
20% Iron-impregnated corn straw biochar [125]	0.2	40		3	87–70	No data	He R. Z. et al., 2018
Modified <i>Saccharum officinarum</i> bagasse (SCB-S) [133]	0.05	0.5		5	100–78	100–86	Gupta A. et al., 2015

6.5. Thermodynamic

The variation in temperature can have positive or negative impacts on the adsorbent's adsorption capacity [134,135]. Usually, for the adsorption thermodynamic study, Gibbs free energy (ΔG°), entropy (ΔS°), and enthalpy (ΔH°) of the adsorption are calculated using van't Hoff thermo-dynamic equations:

$$\Delta G^\circ = -RT \ln(K_D) \quad (21)$$

$$\ln(K_D) = \Delta S^\circ / R - \Delta H^\circ / (RT) \quad (22)$$

where R is the universal gas constant (8.314 J/mol K), T is temperature (K), and K_D (q_e/C_e) is the distribution coefficient. According to Equation (22), the entropy (ΔS°) and enthalpy (ΔH°) of the adsorption parameters can be determined from the slope and intercept of the plot of $\ln(K_D)$ vs. $1/T$ yields, respectively, and then used to calculate the Gibbs free energy (ΔG°) [129].

The term physical sorption means a van der Waals type force formed between the interfaces and chemisorption, which denotes a chemical bond formed between the As molecule and the natural adsorbent surface. Generally, the physical sorption enthalpy (ΔH°) is in the range of -20 to -40 kJ mol^{-1} and chemisorption enthalpy (ΔH°) in the range of -400 to -80 kJ mol^{-1} [136,137]. Usually, the adsorption of As on natural adsorbents are spontaneous chemisorption processes [133]. Take chemically modified watermelon rind for example, the negative values of ΔG° suggested that both As(III) and As(V) adsorption was spontaneous; the positive values of ΔH° for both As(III) (67 kJ mol^{-1}) and As(V) ($86.05 \text{ kJ mol}^{-1}$) sorption indicated that sorption was endothermic; the positive ΔS° values for both As(III) (0.24 kJ mol^{-1}) and As(V) (0.58 kJ mol^{-1}) suggested a disorder at the solid/solution interface along with structural changes [132].

6.6. Influence of Other Ions

Many common ions which can be present in water, especially groundwater, can negatively impact the removal of As due to the capacity of these ions to compete with As for adsorption [138]. In general, as shown in the Table 4, the adsorption capacity of As onto a natural adsorbent in the presence of positive anions (Mg^{2+} , Ca^{2+} , Mn^{2+}) is higher than in the presence of negative anions (NO_3^- , SO_4^{2-} , PO_4^{3-}) because of the electrostatic competition between the negative other anions and the As anions for the adsorption onto the natural adsorbent surface, and the repulsion between the positively charged natural adsorbent surface and the positive anions [89,139,140]. Minerals might have better selectivity compared to biomass, for example, leonardite was used for As removal and the results showed that the enhanced SO_4^{2-} concentration in the solution led to the reduction in As removal capacity from 100 to 95% [141], while Baig et al. [135] found that the removal efficiency of As(III) on the iron modified Kans grass (*Saccharum spontaneum*) biochar dropped notably below 20%, when the concentration of PO_4^{3-} increased to 1.0 mmol/L .

Table 4. Other ions influence on some natural adsorbents.

Adsorbents	Thermodynamics	Kinetic	Influence Ions (mg/L)		Removal As (%)	
					As(III)	As(V)
Acid-activated laterite (AAL) [134]	Endothermic	Pseudo-second-order	PO_4^{3-}	0–10	89–70	95–85
			SO_4^{2-}	0–40	89–68	95–83
Fe_3O_4 -HBC-1000 °C (Honeycomb Briquette Cinders) [142]	Endothermic	Pseudo-second-order	PO_4^{3-}	0–9.5	100–15	100–60
Fe-Mn modified corn stem biochar (FMBC) [143]	Endothermic	Pseudo-second-order	PO_4^{3-}	0–9500	100–88	No data
Iron oxide amended rice husk char (950 IOA-RHC) [138]	Endothermic	Pseudo-second-order	PO_4^{3-}	0–10	No data	100–65
Fe-impregnated hickory chips biochar [144]	No data	No data	PO_4^{3-}	0–50	No data	100–15
Magnetic Kans Grass (<i>Saccharum spontaneum</i>) Biochars (MKGB4) [135]	No data	Pseudo-second-order	PO_4^{3-}	0–95	100–20	100–80
Leonardite chars [141]	No data	No data	SO_4^{2-}	0–5000	98–92	99–94
Siderite SiO_3 [145]	No data	No data	PO_4^{3-}	0–10	No data	64–28
Hematite HIO1 [145]	No data	First-order	PO_4^{3-}	0–10	No data	69–36

6.7. Adsorption Isotherm and Kinetic Model

In the order to design an As removal system on a real scale, studies of adsorption isotherm and kinetics equation models are important to describe the nature of adsorption onto the natural adsorbents, and determine the adsorption capacity of As [146–149]. Many isotherm equation models exist, but the most use ones in the studies of As adsorption are the Langmuir Equation (23) and Freundlich Equation (24) linear equation.

$$C_e/q_e = C_e/q_m + 1/q_m K_a \quad (23)$$

where C_e is the equilibrium concentration, q_e is the equilibrium adsorption capacity of As onto adsorbent, q_m is the maximum adsorption capacity, and K_a is the Langmuir sorption equilibrium constant [150].

$$\log q_e = 1/n \times \log C_e + \log K_f \quad (24)$$

where C_e and q_e have the same meaning as explained above, K_f and n are the Freundlich isotherm constants, representing the adsorption capacity and intensity, respectively [151].

The Langmuir model is an equation which is used to describe monolayer adsorption onto a homogeneous surface. The Freundlich model assumes chemisorption on heterogeneous surface [152,153]. As we can see in Table 1, the Langmuir model is the isotherm sorption model that most of the time explains the adsorption mechanism when a natural adsorbent is used.

There are two kinetics models that are mostly used, which are the pseudo-first-order model (Equation (25) [154] and pseudo-second-order model (Equation (26) [155].

$$\log(Q_e - Q_t) = \log Q_e - k_1/2.303t \quad (25)$$

where Q_e is the amount of As adsorbed on the surface of the adsorbent at equilibrium ($\mu\text{g/g}$), Q_t is amount of As on the surface of the adsorbent at time any t ($\mu\text{g/g}$), and k_1 is the equilibrium rate constant of pseudo-first-order sorption (L/min). The rate constants are calculated by plotting $\log(Q_e - Q_t)$ vs. t .

$$t/Q_t = 1/k_2 Q_e^2 + t/Q_e \quad (26)$$

where t , Q_t , and Q_e have the same meaning as explained above. k_2 is the rate constant of pseudo-second-order adsorption ($\text{g} \cdot \mu\text{g}^{-1} \cdot \text{min}^{-1}$). The rate constants are calculated by plotting t/Q_t vs. t .

The pseudo-second-order kinetic model assumes that chemisorption is the main adsorption mechanism [156] and the pseudo-first-order kinetic model indicates that physical sorption is the adsorption mechanism [140]. Mostly, as we can see in Table 4, when a bio-material, such as the iron-modified chitosan hollow fibers membrane, ZnCl_2 -activated pig manure residue biochar, pine bark, pine wood, oak bark, and oak wood biochar is used for As(III) or As(V) removal, the principal adsorption mechanism is a chemisorption [157–159].

7. Conclusions

Some biosorbents, such as fruit peels (orange, citrus, mango) and minerals (geothite) have a very nice As removal performance, but some others such as biomass or fibers (cotton, sugarcane, coconut) first need a pre-treatment to enhance their performance. As removal by natural adsorbent was found to be mainly endothermic and a spontaneous monolayer chemisorption, and depended on many different parameters; high As initial concentration may reduce the biosorbent As removal effectiveness, but more contact time can help to uptake more As from the water. Enough dosage of the natural sorbent and suitable water flow are very useful for better As removal performance, the adsorption of As(V) is suitable in acidic solution and As(III) is appropriate in a basic solution. The economic factor is the main reason why it will be better to use as natural adsorbent of As from water, a natural material which has no use in our society and usually becomes waste

materials, such as sugar cane fibers, corn fibers, corn cane fibers, groundnut shell. The low cost, good adsorption, and desorption of the biosorbent prove that natural adsorbents are a good As adsorbent, which can help to set up an effective technology for any kind of water polluted by As. In the future, all these advantages of the natural sorbents for As removal from water should be presented to researchers, to increase the research using biosorbents for further As adsorption technology development. Comparisons of the natural sorbent As adsorption capacity with or without modification should be made to reduce the use of chemical products in the process. Moreover, more studies should be conducted using real polluted natural water to check the performance of natural sorbent in real conditions, especially using the column process.

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