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Investigation on *Melia azedarach* biomass for arsenic remediation from contaminated water

Amna Sarwar[a], Qaisar Mahmood[a,*], Muhammad Bilal[a], Zulfiqar Ahmad Bhatti[a], Arshid Pervez[a], Ahmad Nauman Shah Saqib[b], Abdur Rehman Khan[b], Sikander Sultan[c]

[a]*Department of Environmental Sciences, COMSATS Institute of Information Technology, Abbottabad 22060, Pakistan*
Tel. +92 992 383591; Fax: +92 992 383441; email: mahmoodzju@gmail.com
[b]*Department of Chemistry, COMSATS Institute of Information Technology, Abbottabad 22060, Pakistan*
[c]*Department of Microbiology and Molecular Genetics, University of the Punjab, Quaid-i-Azam Campus, Lahore 54590, Pakistan*

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ABSTRACT

The arsenic removal efficiency of *Melia azedarach* biomass was investigated. The effects of pH, biosorbent dose, adsorbate concentration and temperature on the arsenic adsorption were investigated. Batch experiments were conducted using different amounts of biosorbent (2–10 g) at varying amounts of adsorbate (50–500 μg/L) at 20°C. The effect of pH (2–11) on adsorption process was also investigated. The data fit into isotherm models. The study revealed that As(III) and As(V) removal was >90% at an adsorbent dose of 10 g/L, adsorbate concentration of 100 μg/L under pH range of 7–8 for 30 min. Temperature does not significantly affect the As removal efficiency. The experimental data follow Freundlich isotherm. Arsenic adsorption on biomass is confirmed by using scanning electron microscope. This study is pioneer report on Melia biomass for arsenic removal, and it is concluded that being environmentally safe technology, *M. azedarach* biomass can be efficiently utilized for arsenic removal.

Keywords: Arsenic; Biomass; *Melia azedarach*; Biosorbent; Adsorption isotherms

1. Introduction

Arsenic is the 20th most abundant element in nature [1]. It is widely distributed throughout the earth's crust and commonly found in the atmosphere, organisms, rocks, soils and in natural waters. In water reservoirs, its presence may be due to industrial waste discharges, arsenic-containing pesticides, herbicides and fertilizers are responsible for release of As into aquatic environment. Other sources of arsenic include dyestuffs, chemical warfare gases, glass industry, electronics, petroleum refining and ceramic industries [2,3]. A trace amount of arsenic is also present in coal and oil. Arsenic is released into the atmosphere on burning of fossil fuels. Thermal power stations also releases arsenic into ground water sources when the arsenic-containing ash is dumped the in landfills [4].

Arsenic contamination problem is complex because it is tasteless, colourless and odourless imparting chronic effects, and it is not easily detected in water. Contaminated drinking water is one of the main causes of diseases in the developing countries. Arsenic is a lethal contaminant and being a carcinogen, it affects the human health. The permissible limit of arsenic in drinking water is 10 ppb as defined by WHO [4]. Among other symptoms of arsenic toxicity

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*Corresponding author.

in human include skin, lung and liver cancers, diabetes [5], hypertension [6], black foot disease [4], mucous membranes damage, and effects on respiratory, circulatory, digestive and nervous systems [2]. Other diseases that are assumed to be caused or aggravated by drinking arsenic-contaminated water include post-neonatal mortality, still births, developmental discrepancy, heart attack and nephritis [7].

Arsenic contamination is considered to be a global issue due to its toxicity and wide spread prevalence in water sources. Arsenic contamination in ground water resources is observed in many countries; Alaska, Australia, Austria, Bangladesh, Canada, China, France, Germany, Greece, Hungary, India, Iran, Italy, Japan, Mexico, Nepal, New Zealand, Pakistan, Philippines, Romania, Russia, Taiwan, Thailand, UK and USA [2,4,8,9]. About 35 million people are affected by arsenic contamination in India, Nepal and Bangladesh [10].

Several methods and techniques have been developed for the arsenic removal from water and wastewater that includes oxidation, coagulation, co-precipitation, ion exchange, adsorption and membrane techniques [11]. Coagulation is the most commonly used technique but sludge disposal is a major concern. Ion-exchange resins may result in fouling and regeneration is required at definite time intervals. Membrane technology is very expensive and is not applicable on small scale [12].

Melia azedarach is a tall deciduous tree belonging to family; Meliaceae. This tree is fast-growing wild tree and native to India, China, Australia and also found in Pakistan. The adult tree has rounded crown and commonly attain a height of 7–12 m; it can also attain the height of 45 m in exceptional circumstances. The leaves are up to 50 cm long. In past few decades, *M. azedarach* has been used to treat skin diseases in addition to pesticides and insecticides [13]. Recently, *M. azedarach* was used in the reclamation of soils affected by tannery effluents [14]. In view of prevalence of *M. azedarach* in the tropical areas of Pakistan and Abbottabad, the dry leaves and bark of the plant were used to treat arsenic-containing water.

2. Methods

2.1. Reagents and solutions

Plasticware used (to avoid metal contaminations) was cleaned in hydrochloric acid (20%, v/v) followed by nitric acid (20%, v/v) (Merck, Darmstadt, Germany) and rinsed with distilled deionized water. All the reagents used in the experiment were of analytical grade and were purchased from representative of Sigma–Aldrich in Pakistan.

Stock solution of As (1,000 μg/mL) was prepared from atomic absorption standard (Perkins Elmer, USA). Other standards were prepared by dilution of the stock in UHP water. Hydrochloric acid and NaOH (0.1 M solutions) were used for pH adjustments.

2.2. Adsorbent collection and storage

The biomass was collected from the university campus (CIIT Abbottabad). After collection, the biomass (leaves and bark) was subjected to dry in shade for a period of three weeks. After drying, it was ground to reduce its size using pestle and mortar before washing with distilled water to remove dust, colour and other impurities. After washing, it was oven-dried at 70˚C. The washed and dried biomass was then stored in a desiccators till further use.

2.3. Instrumentation and characterization of adsorbent material

Arsenic analysis was carried out by the standard colorimetric method reported by Bigham using UV–vis Spectrophotometer at 840 nm [15]. To develop calibration curve, 0.5, 1.0, 2.0, 3.0 … 8 ml of standard arsenic solutions (AsO_3) were added into respective 50 ml of conical flasks, and then, 2 ml of acid molybdate solution and 7 ml of sulphuric acid was added. The solution was mixed and heated the flask on hot plate until solution was reduced to 5–6 ml. The solution was allowed to cool and to each added 1 ml of 0.030% w/v solution of hydrazine sulphate in water, mixed and solution was drained into the 10 ml of cylinder, flask was rinsed with small amount of water and washing was used to dilute the main solution to the 10 ml. The solution was thoroughly mixed and returned each solution to its original 50 ml conical flask. The mouth of the flask was closed with glass bulb, and flask was heated on the boiling water bath for fifteen minutes. The flask was removed from the water bath and allowed to cool for thirty minutes. The absorbance of the prepared standard solutions was measured at 840 nm against blank solution, prepared in the similar manner without the arsenic. A graph was constructed between the absorption values to the number of microgram of arsenic.

Scanning electron microscope (SEM) was used to characterize the surface of adsorbent materials. Scanning electron micrographs of the adsorbent material (unloaded and arsenic loaded) were studied at the magnification, i.e. ×20 mm for leaves and ×1.0 mm for bark.

2.4. Biosorption process

Effects of various parameters (pH, biosorbent dose, biosorption, adsorbate concentration and temperature) were investigated on the As adsorption by Melia biomass. Batch experiments were conducted to study As adsorption using different amounts of biosorbent material (2–10 g) with varying amounts of adsorbate (50–500 μg/L) at 20˚C. The pH (2–11) was adjusted using 0.1 M HCl or 0.1 M NaOH. The time required to reach sorption equilibrium was estimated by analysing the adsorbate at regular time intervals. After sufficient time period, the biomass was removed through filtration, washed with UHP water twice, and the resulting solution was analysed. The arsenic concentration at all intervals of time was determined. All experiments were conducted in triplicate, and the mean values were used in all calculations. The per cent adsorption of the biosorbent was calculated by using the following formula:

$$\%\text{Sorption} = C_i - C_e/C_i \times 100 \tag{1}$$

where C_i and C_e are the initial and equilibrium concentrations of arsenic expressed in μg/L.

3. Results

3.1. Effect of adsorbent dose

The effect of adsorbent doses (2–10 g) on the As removal efficiency was studied. Adsorbate dose was fixed (100 μg/L), and a contact time of 30 min was for the reaction. The samples were filtered, and arsenic was removed. The study revealed that the adsorption of As(V) was greater (97%) that that of As(III) (60%) with the increasing concentration of bark as adsorbent (Fig. 1(a)). A similar trend was noted when the Melia leaf powder was used as adsorbent (Fig. 2(a)). Further increase in the amounts of adsorbent has no obvious effect on As removal efficiency. This effect is obvious, as the increase in the amount of adsorbent causes an increase in surface area which in turn increases the removal efficiency.

3.2. Effect of adsorbate dose

In next phase, the effect of adsorbate dose (50–500 μg/L) on arsenic removal efficiency was investigated keeping the adsorbent dose fixed (10 g). In case of bark, an increase in the removal efficiency was observed as adsorbate dose was increased (50–200 μg/L). At 200 μg/L, more than 90% of As(III) and As(V) were adsorbed on the biomass. Further increase in the adsorbate dose (up to 500 μg/L) did not affect the removal efficiency (Fig. 1(b)). In case of leaves, As(III) adsorption efficiency of 91–93% was observed for adsorbate concentration of 50–100 μg/L and efficiency decreased from 85 to 65% when adsorbate concentration was increased to 500 μg/L. The adsorption efficiency of As(V) decreases from 98% at 50–100 μg/L to 65% at adsorbate concentration of 500 μg/L (Fig. 2(b)). It is implied that the sorptive surface area available to metal ion is high at very low concentration of metal ion; therefore, greater chance for metal removal at that adsorbent dose. Thus, there is higher removal capacity at low metal ion concentration. As the metal concentration increases, the binding sites become saturated if the amount of biomass concentration remains constant.

3.3. Effect of pH

Keeping adsorbate dose (100 μg/L), adsorbent (10 g) and contact time (30 min) constant, the effects of pH on the As adsorption was investigated in the range of 2–11. The results showed that the As adsorption increased with the increasing pH (2–11) for both bark and leaves biomass. The maximum As adsorption (93–98%) on both biomasses was observed at slightly alkaline pH (8) (Figs. 1(c) and 2(c)).

3.4. Effect of contact time

The dependency of As removal on contact time was also investigated at various time intervals (10–80 min), fixed adsorbent dose (10 g), adsorbate concentration (100 μg/L) and optimum pH (8). This study also revealed that the maximum removal of As took place during contact time period of 20–30 min, for both bark and leaves biomass (Figs. 1(d) and 2(d)). Arsenic adsorption was fast within first 30 min suggesting the occupation of active sites on biomasses and more than 90% removal takes place during that period. After the process reached at steady state, As adsorption percentage was not affected by increase in contact time.

3.5. Effect of temperature

Arsenic adsorption was also investigated at various temperatures (5–45˚C) using fixed adsorbent dose (10 g), adsorbate concentration (100 μg/L), pH (8) for 30 min. The temperature was controlled using a water bath. The results showed that there was no significant effect of temperature on the adsorption process using various adsorbent materials (Figs. 1(e) and 2(e)).

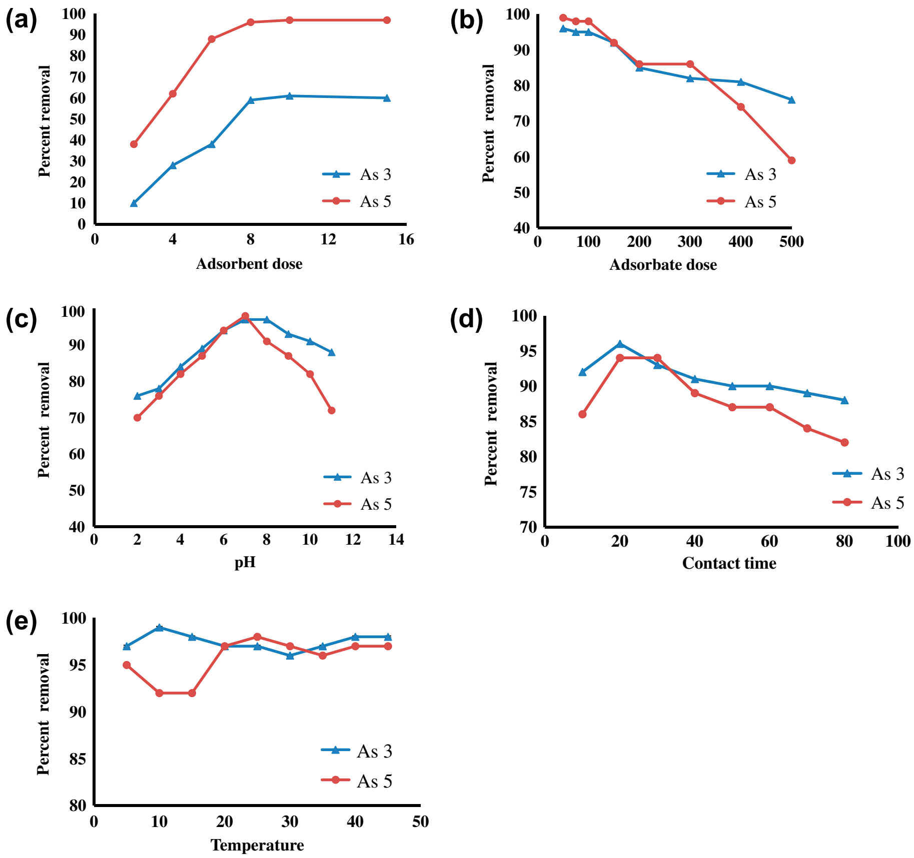

Fig. 1. (a) The effect of adsorbent dose on the removal of As(III) and As(V) using Bark Biomass. (b) The effect of adsorbate dose on the removal of As(III) and As(V) using Bark Biomass as adsorbent. (c) The effect of pH on the removal efficiency of As(III) and As(V) using Bark Biomass as adsorbent. (d) The effect of contact time on the removal efficiency of As(III) and As(V) using Bark Biomass as adsorbent. (e) The effect of temperature on the removal efficiency of As(III) and As(V) using Bark Biomass as adsorbent.

3.6. Scanning electron micrograph of leaves

Scanning electron micrographs were taken to study the topography of the loaded and unloaded Melia biomass. Under the optimized conditions for As biosorption, the loaded and unloaded biomass were subjected to the arsenic solutions prior to SEM analysis, in order to check the changes on the surface of biomass before and after the reaction. The scanning electron micrographs were taken at resolution of ×20 mm for leaves biomass and ×1.0 mm for bark biomass. Figs. 3(a) and (c) show rough surface with pores that possibly act as sites for As biosorption either physically or chemically. The presence of large particles on the adsorbent surface is clear in Figs. 3(b) and (d). SEM studies indicated that densities of the particles of the treated leaves and bark were higher than the untreated samples supporting the fact that aresnic particles adsorbed onto the surface of leaves and bark of *M. azedarach*.

3.7. Adsorption isotherms

Adsorption isotherms are the mathematical models that throw light on the amount of the adsorbate distributed between liquid and adsorbent, the amount of adsorbate adsorbed by adsorbent and type of interaction between the adsorbate species with adsorbent. These models describe the equilibrium of the sorption of a material at a surface at constant temperature. In this study, Langmuir and Freundlich isotherms were applied to the experimental data (Table 1).

The Langmuir model is based on the assumption that the maximum adsorption occurs when a saturated monolayer of solute molecules is formed on the adsorbent surface, and there is no migration of adsorbate molecules in the surface area and a single solute binds to specific homogenous sites [16].

Langmuir isotherm is given by:

$$1/q = 1/q_m K_{\mathrm{ads}}(1/C) + 1/q_m \tag{2}$$

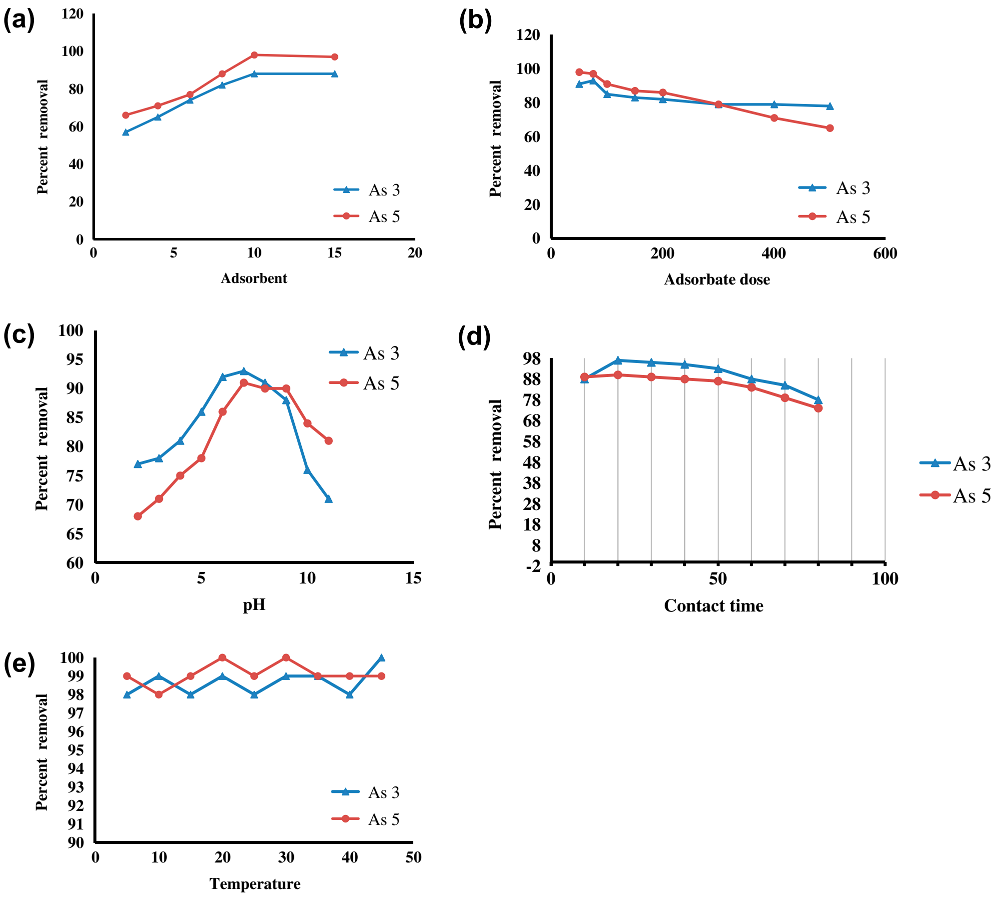

Fig. 2. (a) The effect of adsorbent dose on the removal efficiency of As(III) and As(V) using Leaves as adsorbent. (b) The effect of Adsorbate on the removal efficiency of As(III) and As(V) using Leaves as adsorbent. (c) The effect of pH on the removal efficiency of As(III) and As(V) by using Leaves as adsorbent. (d) The effect of contact time on the removal efficiency of As(III) and As(V) using Leaves as adsorbent. (e) The effect of temperature on the removal efficiency of As(III) and As(V) using Leaves as adsorbent.

where q is the adsorbed concentration (mass of adsorbate/mass of adsorbent), q_m the maximum capacity of adsorbent for the adsorbate (mass of adsorbate/mass of adsorbent), C the initial concentration of adsorbate in solution (mass per unit volume) K_{ads} the adsorbate affinity for adsorbent. A plot of $1/q$ against $1/C$ produced a straight line with an intercept of $1/q_m$ and a slope of $1/q_m.K_{\text{ads}}$. R^2 values of 0.554 and 0.614 for As(III) and As(V), respectively, for bark biomass and R^2 values of 0.774 and 0.614 for As(III) and As(V), respectively, for leaves biomass revealed that the adsorption of arsenic by adsorbent did not follow Langmuir isotherm.

The Freundlich isotherm is a mathematical relationship that describes the adsorption of solute particles from a solution to a solid surface, and it is assumed that they have different affinities for different adsorbates. Expression of Freundlich isotherm is as follows:

$$q = KC^{1/n} \tag{3}$$

where K is the measure of the capacity of adsorbent, and n is the measure of change of affinity for the adsorbate with a change in adsorption density.

If $n = 1$, then it means that all the active sites present on adsorbent surface have the same affinity for the adsorbate and results in linear isotherm. But if value of n is greater than 1, it shows that affinity decreases for the adsorbate with an increase in adsorption density.

$$\text{Log } q = \log K + 1/n \log C \tag{4}$$

A plot of log q against log C produced a straight line with a slope of $1/n$ and an intercept of log k. R^2 values of 0.99 and 0.97 for As(III) and As(V), respectively, for bark biomass and R^2 values of 0.99

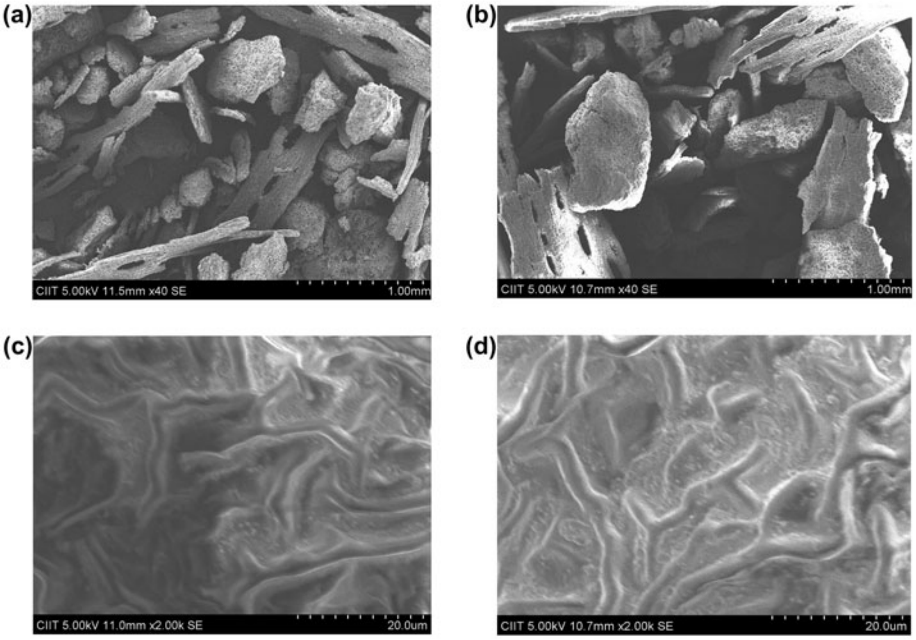

Fig. 3. (a) Scanning electron micrograph of unloaded Bark of *M. azedarach*. (b) Scanning electron micrograph of loaded Bark of *M. azedarach*. (c) Micrograph of unloaded leaves powder of *M. azedarach*. (d) Scanning electron micrograph of loaded leaves powder of *M. azedarach*.

Table 1
Langmuir and Freundlich isotherms

	Bark biomass		Leaves biomass	
Adsorbate	Langmuir isotherm (R^2) value	Freundlich isotherm (R^2) value	Langmuir isotherm (R^2) value	Freundlich isotherm (R^2) value
As(III)	0.55	0.99	0.77	0.99
As(V)	0.61	0.97	0.61	0.96

and 0.96 for As(III) and As(V), respectively, for leaves showed that the data were in great agreement with Freundlich isotherm.

4. Discussion

The adsorption method is very attractive for its easy handling, minimal sludge production and its regeneration capability. A number of adsorptive media have been investigated for the removal of arsenic from drinking water. Various investigators reported As(III) and As(V) from environmental samples and demonstrated the effectiveness of various biomasses to remediate aqueous media [17–20]. Some of those are: activated carbon, activated alumina, iron and manganese coated sand, activated bauxite, kaolinite clay, hydrated ferric oxide, titanium oxide, phyllosilicates, silica, and hydrous oxides of Fe and Al and many natural and synthetic media [9,11,21–23]. Arsenic adsorption is found to be more dependent on its oxidation state rather than pH within the pH range of 5.5–7.5 [24]. However, the optimum pH for As adsorption on Melia biomass during the current study was found as slightly alkaline (8). An increased attention is being paid to the biosorbents for the removal of arsenic and other metals from water and wastewaters [25]. Various low-cost biomaterials include plant biomass, chitin and chitosan, algae, fungi and bacteria. Powdered biomass of *Acacia nilotica* has been reported to be quite efficient in arsenic removal [2]. Some other biomaterials derived from fish scales, coconut fibre, dried roots of water hyacinth plant, seed powder of *Moringa oleifera*, *Momordica charantia*, on lichen (*Xanthoria parietina*), powdered eggshell, human hair, rice husk, rice polish; without chemical treatment have been reported [4,26–33]. However, still there is a need to develop cost-effective and commonly available biosorbent for arsenic removal.

Recently, arsenic adsorption using natural materials or the wastes products from industrial or agricultural operations has emerged as an option for developing economic and eco-friendly wastewater treatment processes. Numerous low-cost adsorbents have so far been studied for the removal of arsenic from water and wastewater. Dry plants, red mud, fly ash, zeolites, blast furnace slags, hydrotalcites, hydroxides and various bioadsorbents are few to mention. The removal of heavy metals by these materials might be attributed to their protein, carbohydrates and phenolic contents, which have metal-binding functional groups such as carboxyl, hydroxyl, sulphate, arsenate and amino groups. Mohan and Pittman [34] observed that the loss of metal ions from their solutions in the presence of natural materials may be due to the adsorption on surface and pores and to complexation of these materials.

Dried plants are natural materials widely available and studied as an alternative adsorbent for different heavy metals. Examples of proposed plant leaves as efficient in removing metal ions from water are reed [35] for cadmium, poplar for lead and copper [35,36], cinchona for copper and lead [36,37], pine for cadmium and nickel [38,39] and cypress for aluminium [39], Nile rose plant powder for lead ions [40,41]. Abdel-Halim et al. [41] reported the removal of zinc from aqueous solutions by 15 species of plant leaves. They found that the removal efficiency of zinc dependent on the plant species used. Dry plant leaves of thyme, sage, banana, mint, anise and oleander plants have also been suggested as natural, simple and cheap adsorbent for the efficient removal of several metal ions from polluted water [42]. It was demonstrated that crushed dried plants such as *Asphodelus microcarpus*, *Asparagus albus*, *Carpobrotus edulis*, *Euphorbia echinus*, *Launea arborescens*, *Senecio anthophorbium* and *Withania frutescens* which grow in the Agadir area could act as adsorbents of pollutants such as arsenate with high efficiency [43–47]. Romanian *Cyperus rhizome* plant was also used for arsenate removal from aqueous solution. The results showed that the maximum adsorption capacity of arsenate on *C. rhizome* plant was 22.04 mg/g at initial arsenate concentration of 1 g/L [48]. The biomass of *A. nilotica* was found to be effective for the removal of As with 95% sorption efficiency at a concentration of < 200 mg/L of As solution, and thus, uptake capacity is 50.8 mg As/g of biomass [2]. In this study, the Melia biomass had relatively lower adsorption capacity of 15–25 mg As/g of biomass. The biosorption capacity of *I. hispidus* for As(III) and As(V) was found to be 51.9 and 59.6 mg/g, respectively, at optimum conditions of pH 6 for As(III) and pH 2 for As(V), contact time of 30 min and temperature of 20°C [17]. The biosorption capacity of *Ulothrix cylindricum* biomass was found as 67.2 mg/g [18]. It has also been shown that the order of the fixation efficiency depended on the plant used. Moreover, the efficiency of arsenate adsorption depends on different parameters (size of crushed plants which must be less than 50 μm, m/V of crushed vegetal mass and volume of solution to be treated, solution concentration and pH). It is noteworthy that vegetative materials represent a potential source of abundant low-cost adsorbents, and there is no environmental or technical reason which impedes the preparation of adsorbents by using those materials [49,50].

5. Conclusion

The *M. azedarach* biomass was found capable of >90% removal of both Arsenite (As(III)) and Arsenate (As(V)) from the aqueous solutions. Temperature does not affect the arsenic removal efficiency. SEM confirmed the topographical changes on the sorbent surface caused by arsenic adsorption. The results were found to obey the Freundlich isotherm for both As(III) and As(V). The proposed adsorbent is suitable and easily applicable in local areas because of its simplicity, easy operation and handling and cost-effectiveness. It can be used for the arsenic removal from drinking water in the range of 100–300 μg/L.

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