

Research Article

Arsenic Removal from Aqueous Solutions by *Salvadora persica* Stem Ash

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Arsenic is a naturally occurring metalloid, which is widely distributed in nature and is regarded as the largest mass poisoning in history. In the present study, the adsorption potential of *Salvadora persica* (*S. persica*) stem ash in a batch system for the removal of As(V) from aqueous solutions was investigated. Isotherm studies were carried out to evaluate the effect of contact time (20–240 min), pH (2–11), initial arsenic concentration (50–500 µg/L), and adsorbent dose on sorption efficiency. Maximum removal efficiency of 98.33% and 99.32% was obtained at pH 6, adsorbent dosage 3.5 g/L, initial As(V) concentration 500 µg/L, and contact time 80 and 60 min for *S. persica* stem ash at 300 °C and 500 °C, respectively. Also, the adsorption equilibriums were analyzed by the Langmuir and Freundlich isotherm models. Such equilibriums showed that the adsorption data was well fitted with the Freundlich isotherm model for *S. persica* stem ash at both 300 °C and 500 °C ($R^2 = 0.8983$ and 0.9274, resp.). According to achieved results, it was defined that *S. persica* stem ash can be used effectively for As(V) removal from the aqueous environment.

1. Introduction

Arsenic (a semimetallic element) is one of the most toxic, carcinogenic, and natural contaminants found in water resources [1, 2]. Arsenic in water systems is mostly introduced from natural and anthropogenic sources. Natural sources include the washout and erosion of arsenic-rich rocks and soils, which probably occur because of long-term geochemical changes. Anthropogenic sources include forestry, agricultural application of various pesticides, herbicides and fertilizers, and industrial effluents from metallurgy, electronics, mining, pharmaceuticals, glass processing, ceramic, dye and pesticides manufacturing, wood preservatives, petroleum refining, and landfill leaching [3, 4].

The two states prevalent in water environment are arsenite (As(III)) and arsenate (As(V)) [5]. As(III) mostly exists in reducing ground waters and hydrothermal waters, whilst As(V) is more often present in surface waters and oxidizing ground waters [6]. Arsenic is a geogenic water menace affecting millions of people all over the world and is regarded

as the largest mass poisoning in history. Permanent arsenic intake can lead to chronic intoxication, and prolonged arsenic exposure can damage the central nervous system, liver, and skin and results in the appearance of diverse types of cancers, such as hyperkeratosis, lung, skin, and prostate cancers [7–9].

Arsenic in natural waters is a worldwide problem. Arsenic pollution has been reported recently in USA, China, Chile, Bangladesh, Taiwan, Mexico, Argentina, Poland, Canada, Hungary, New Zealand, Japan, and India [10]. The largest population at risk among the 21 countries with known groundwater arsenic contamination is in Bangladesh, followed by West Bengal in India [10]. Due to high toxicity and carcinogenic effect of arsenic, the World Health Organization (WHO), the European Commission (EU), and the United States Environmental Protection Agency (USEPA) have recommended a maximum contaminant level (MCL) of 10 µg/L for arsenic in drinking water [11]. In Iran, a maximum arsenic level of 10 µg/L is permitted in drinking water by the Institute of Standard and Industrial Research of Iran (ISIRI).

Arsenic removal from contaminated water is an important subject worldwide, which has recently attracted great attentions. Several treatment technologies have been applied for the removal of arsenic from water, including chemical coagulation [12, 13], chemical oxidation [14], dissolved air flotation (DAF) process [15], adsorption [3], ion exchange [16], membrane processes [17], electrocoagulation [18], constructed wetlands [19], and biological process [20]. Each method has its advantages and disadvantages, and individual water treatment plants may choose the method that best suits their financial and managerial situations.

Coagulation and adsorption processes are most promising for arsenic removal from high-arsenic water because of the low cost and high efficiency and are widely used in the developing world [21]. Although the coagulation process is a simple and economical way, it produces a wet bulky sludge, and furthermore could not satisfy the stringent standard of the WHO. Because of the ease of handling, sludge-free operation, and using acid or base leaching to regenerate the sorbent bed, the adsorption process appears to be the most promising one [22]. Also, currently, low-cost adsorption materials are popular for arsenic removal from natural water [23, 24]. On the other hand, adsorption has a comparatively low cost, availability, and easy operation and it easily separates a small amount of toxic elements from large volumes of solutions [25]. These advantages of adsorption have motivated several researchers to use this process for removal of arsenic from water. Some of the common adsorbents used for arsenic removal include activated alumina, activated carbon composites, granular ferric hydroxide, manganese green sand, natural laterite, rare earth oxides, bone char, surface-modified zeolite with cationic surfactant and mud, and Fe-exchanged natural zeolite [22, 26–30].

Salvadora persica or Arak is an evergreen perennial halophyte belonging to the family Salvadoraceae, and it is considered an important crop plant for marginal coastal areas of arid and semiarid regions, where the salt concentration of the soil would inhibit the growth of most other crops [31, 32]. *S. persica* can tolerate salinity up to 635 mM, and it performs well in temperatures up to 45°C in presence of annual rainfalls varying from 200 to 1000 mm. Moreover, it exhibits notable flexibility since it can be cultivated in saline and nonsaline soils with various textures from sandy to clay soils. Therefore, it can be considered a drought- and salt-tolerant species, in addition to its ecological function in counteracting the advancement of desertification as an effective soil binder [31].

S. persica has a wide geographic distribution ranging from Rajasthan (India), Nepal, and Malaysia in the east through Pakistan, Iran, Iraq, Saudi Arabia, and Egypt to Mauritania in the west, and from North Africa through Sudan, Ethiopia, and Central Africa to southwestern Africa [33, 34].

The main objective of this research was to evaluate the feasibility of using the *S. persica* stem ash as an adsorbent for the removal of arsenic from aqueous solutions. The effects of different parameters including solution pH, adsorbent dosage, arsenic concentration, temperature of ash produced, and contact time were studied. Additionally, the sorption isotherm was explored to describe the experimental data.

2. Experimental

2.1. Adsorbent Preparation. The stems of *S. persica* were obtained from Arabic commercial sources. These materials were firstly washed with distilled water to remove impurity such as sand and leaves and soluble and colored components, dried at 110°C for 12 h, burned at 300°C and 500°C for 2 h, crushed in a domestic grinder, and sieved to obtain a particle size in the range of 30–100 mesh. The powdered adsorbent was stored in an airtight container until use. No other chemical or physical treatments were used prior to adsorption experiments.

2.2. Arsenic Solution (Absorbate). All the chemicals used were of analytical reagent (AR) grade. Stock solutions of As(V) were prepared by dissolving sodium salt heptahydrate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) in double-distilled water (at a concentration of 10 mg/L) and the experimental solutions were obtained by diluting the stock solution to the required concentrations (50–500 µg/L). The solution pH (2–11) was adjusted with 0.1 M HCl or 0.1 M NaOH.

2.3. Analysis. The residual arsenic concentration in the solutions was analyzed by silver diethyl dithiocarbamate method (SDDC) using UV-Vis spectrophotometer (GBC, Cintra 202) at 530 nm. The pH of solution was measured using a pH meter (Denver Ultra basic-UB10).

2.4. Batch Adsorption Experiments. All experiments were carried out with the adsorbent sample in 100 mL conical flasks containing As(V) solutions to evaluate the experimental parameters such as pH, adsorbent dosage and mesh, arsenic concentration, and contact time. The ranges employed for these parameters are given in Table 1. Following a systematic process, the removal of As(V) from aqueous solutions by the use of *S. persica* stem ash in a batch system was studied in the present research work. The amount of sorption, q (µg/g), was calculated by

$$q_e = \frac{(C_0 - C_e)V}{M}, \quad (1)$$

where C_0 and C_e are the initial and equilibrium concentrations of As(V) (µg/L), respectively, V is the volume of the solution (L), and M is the amount of adsorbent used (g). To express the percent of As(V) removal, the following equation was used:

$$\% = \frac{(C_i - C_f)}{C_i} \cdot 100, \quad (2)$$

where C_i and C_f represent the initial and final (after adsorption) As(V) concentrations, respectively.

2.4.1. Effect of Solution pH on As(V) Adsorption. The effect of solution pH on the adsorption capacity of *S. persica* stem ash (0.35 g) obtained at 300 and 500°C was investigated using a 100 mL solution of 150 µg/L of As(V) for a pH range of 2 to 11 at 23±2°C. Flasks were agitated on a rotary shaker at 190 rpm

TABLE 1: The ranges of experimental parameters.

pH	Adsorbent dosage (g/L)	<i>Salvadora persica</i> stem ash (°C)	Arsenic concentration, µg/L	Contact time, min
2, 3, 4, 5, 6, 7, 8, 9, 10, 11	0.5, 1.5, 2.5, 3.5, 5, 6	300, 500	50, 100, 150, 200, 400, 500	20, 40, 60, 80, 100, 120, 180, 240

for 80 min to ensure that the equilibrium was reached. The mixtures were then filtered through Whatman 42 filter paper and the concentration of As(V) in the filtrates was analyzed using a spectrophotometer. The above procedure was repeated three times and the average value was taken.

2.4.2. Effect of *S. persica* Stem Ash Dosage. Batch adsorption experiments were done at a different *S. persica* stem ash (obtained at 300 and 500°C) concentration from 0.05 g to 0.6 g in a 100 mL solution of 150 µg/L of As(V) at pH 6.0, for a contact time of 80 min at 23 ± 2°C. The samples were then agitated and filtered and the filtrates were analyzed as mentioned before.

2.4.3. Effect of Contact Time. Batch adsorption experiments were carried out at different contact times (20, 40, 60, 80, 100, 120, 180, 240 min) for an initial concentration of 150 µg/L of As(V) solution at pH 6.0. The *S. persica* stem ash (obtained at 300 and 500°C) dose was 0.35 g in 100 mL solution in 250 mL conical flask at 23 ± 2°C. The samples were then agitated and filtered. The filtrates were analyzed as mentioned before.

2.4.4. Effect of Initial As(V) Concentration. Batch experiments were carried out by contacting 0.35 g of *S. persica* stem ash (obtained at 300 and 500°C) with 100 mL of As(V) solution of different concentrations (50, 100, 150, 200, 400, 500 µg/L) at pH value of 6.0 at 23 ± 2°C. A series of such conical flasks were shaken for 60 and 80 min with *S. persica* stem ash at a speed of 190 rpm. Samples were then agitated and filtered. The filtrates were analyzed as mentioned before.

3. Results and Discussion

3.1. Effect of Initial pH. The pH of the solution is one of the most critical parameters in the adsorption of pollutants from aqueous solutions. In order to determine the preferred pH for adsorption of As(V) over *S. persica* stem ash, the uptake of As(V) as a function of hydrogen ion concentration was studied. Figure 1 depicts the effect of pH on adsorption of As(V) onto the *S. persica* stem ash (300°C and 500°C). Both adsorption efficiency and capacity have the same trend. The maximum adsorption capacities of *S. persica* stem ash for As(V) occurred at pH 4.0–9.0. Nevertheless, highest adsorption has taken place at pH 6.0 (95.3%, 40.85 mg/g and 98.6%, 42.24 mg/g for ash 300°C and 500°C, resp.) which was chosen as an optimum pH condition for further experiments. pH parameter is very important since it strictly depends upon the nature of adsorbate and adsorbent. Also, it is an established fact that As(V) is more strongly adsorbed than

As(III) where pH is acidic or near neutral [35]. In addition, after adsorption, the pH of solution was slightly elevated. One reason for the change of pH may be the ion exchange process.

3.2. Effect of Contact Time. The contact time is one of the most important parameters for practical application. The effect of contact time onto the adsorption of arsenic by *S. persica* stem ash at 300°C and 500°C is shown in Figure 2. The adsorption capacity and removal efficiency of As(V) onto the *S. persica* stem ash significantly increase during the initial adsorption stage (0–20 min) and then continue to increase at a relatively slow speed with contact time until a state of equilibrium is attained after 80 and 60 min for ash at 300°C and 500°C. Because the adsorbate is adsorbed easily on the surface (or macropore surface) of the adsorbent, rapid adsorption results. In contrast, in the interior (or micropore surface) of the adsorbent, the adsorbate would be adsorbed by a pore and/or surface diffusion mechanism, resulting in a slower adsorption. On the other hand, according to Figure 2, As(V) was rapidly adsorbed in the first 20 min (77.42% and 80.75% for ash at 300°C and 500°C, resp.), then the adsorption rate decreased gradually from 20 to 240 min, and finally reached equilibrium in about 80 and 60 min. Also, the maximum efficiency of 96.96% and 98.66% for removal of As(V) was obtained at approximately 80 and 60 min for ash at 300°C and 500°C, respectively.

A further increase in contact time had an insignificant effect on the amount of adsorption (adsorption capacity). Therefore, contact time of 80 and 60 min was used in subsequent experiments (for ash 300°C and 500°C, resp.). Generally the removal rate of sorbate is rapid initially, but it gradually decreases with time until it reaches equilibrium. This phenomenon is attributed to the fact that a large number of vacant surface sites are available for adsorption at the initial stage, and after a lapse of time, the remaining vacant surface sites are difficult to be occupied due to repulsive forces between the solute molecules on the solid and bulk phases. Similar findings were reported by other researchers [36–38].

3.3. Effect of Adsorbent Dose. The *S. persica* stem ash dose is an important parameter because this determines the capacity of the adsorbent for a given initial As(V) ion concentration. Therefore, the effect of adsorbent dose on As(V) uptake was depicted in Figure 3, which showed that the adsorption efficiency of As(V) increased very quickly with an increase in dosage of *S. persica* stem ash from 0.5 to 3.5 g/L for both ash 300°C and 500°C; a marginal increased was observed on further increase in the adsorbent dose. On the other hand,

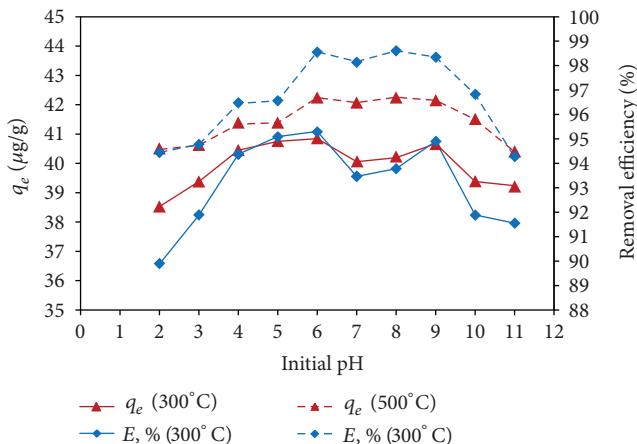


FIGURE 1: Effect of pH for the adsorption of As(V) onto *S. persica* stem ash (the initial As(V) concentration = $150 \mu\text{g/L}$, adsorbent dose = 3.5 g/L , and contact time = 80 min).

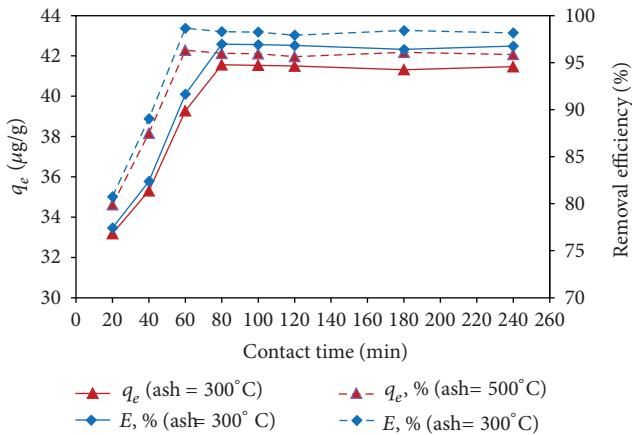


FIGURE 2: Effect of contact time for the adsorption of As(V) onto *S. persica* stem ash (the initial As(V) concentration = $150 \mu\text{g L}^{-1}$, pH = 6, and adsorbent dose = 3.5 g/L).

at lower adsorbent doses, the increase in the percentage of As(V) removal was very high because of the increase in adsorbent dose. However, at higher adsorbent doses, the increase in the removal efficiency of As(V) was insignificant. This may occur because arsenic adsorption happens in two stages, which are fast formation of a monolayer followed by a slow plateau stage [39].

Furthermore, both adsorption efficiency and capacity have the same trend. As shown in Figure 3, adsorption efficiency has gone up from 75.42% and 84.09% at a dose of 0.5 g/L to 96.76 and 98.54% at 6 g/L with a peak at 3.5 g/L (97.21% and 98.57%) for ash at 300°C and 500°C , respectively. Increase in removal efficiency was because of the greater availability of the exchangeable sites or surface area at the higher concentrations of the adsorbent [40, 41]. On the other hand, the increase in the efficiency of removal may be attributed to the fact that with an increase in the adsorbent dose, more adsorbent surface or more adsorption spots were available for the solute to be adsorbed. The results of this study

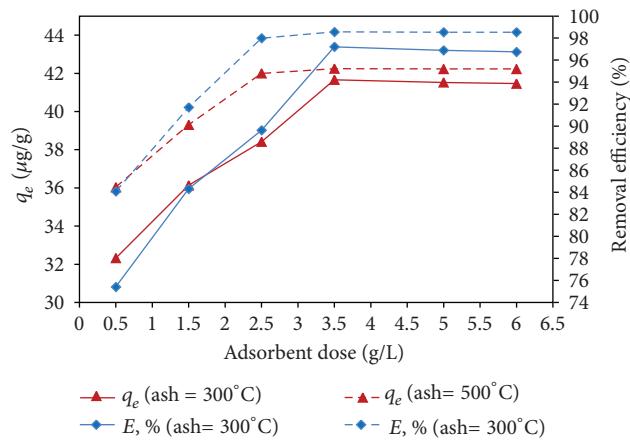


FIGURE 3: Effect of adsorbent dose on As(V) removal (the initial As(V) concentration = $150 \mu\text{g/L}$, pH = 6, and contact time of 80 and 60 min for ash at 300°C and 500°C).

are in accordance with obtained findings by other researchers [3, 38].

The rate of adsorption depends upon the uncovered surface available for adsorption. Initially, as the whole surface was uncovered, the rate of adsorption was very high. As the surface was covered increasingly, the rate of adsorption decreased. Ultimately, a stage was reached when there was no more adsorption with the further addition of adsorbent, and at that time, equilibrium was achieved. The trend in adsorption capacity was because of the interference between binding sites, and higher biomass dose or insufficiency of metal ions in solution with respect to available binding sites [42, 43]. After studying the effect of concentration of adsorbent on ash at both 300°C and 500°C , a dose of 3.5 g/L was selected for subsequent experiments.

3.4. Effect of Initial Concentration of Arsenic. The initial concentration provides an important driving force to overcome all mass transfer resistance of the adsorbate between the aqueous and solid phases [44]. In this study, the effect of initial As(V) concentrations on adsorption process was studied at pH 6, adsorbent concentration 3.5 g/L , and contact time 80 and 60 min for ash at both 300°C and 500°C . As can be seen from Figure 4, when the initial As(V) concentration is increased, the amounts of adsorbed As(V) also increase, so the removal of As(V) depends on the concentration of As(V). On the other hand, the removal efficiency of As(V) by *S. persica* stem ash increased sharply with the increase in the initial As(V) concentration up to $200 \mu\text{g/L}$. Finally, a stage was reached when there was no more adsorption with the more increase of initial As(V) concentration, and at that time, equilibrium was achieved. For example, when the initial As(V) concentration increases from 50 to $500 \mu\text{g/L}$, the equilibrium sorption capacities of *S. persica* stem ash increase from 5.18 and $5.89 \mu\text{g/g}$ (36.26% and 41.26%) to 140.47 and $141.89 \mu\text{g/g}$ (98.33% and 99.32%) for ash at 300°C and 500°C , respectively. Similar trends have also been observed for removal of As(V) from aqueous solutions [3], methylene

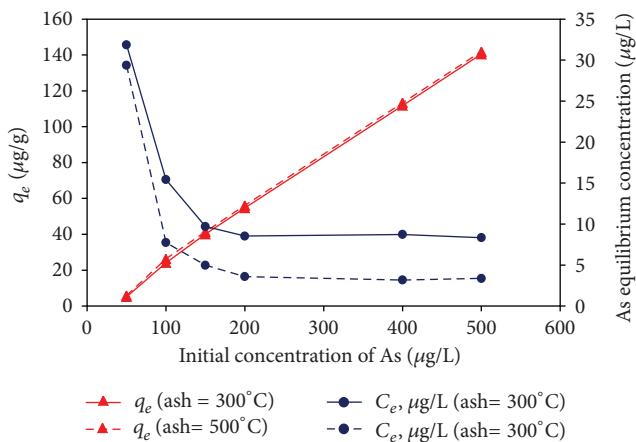


FIGURE 4: Effect of initial As(V) concentration for the adsorption of As(V) onto adsorbent in optimum condition (adsorbent dose = 3.5 g/L, pH = 6, and contact time 80 and 60 min for ash at 300°C and 500°C).

blue dye sorption onto bamboo-based activated carbon [45], cotton waste [46], and Reactive Red 120 removal by single-walled carbon nanotubes from aqueous environments [47]. As mentioned above, the initial sorbate concentrations provide an important driving force to overcome the mass transfer resistance of the adsorbate between the aqueous phases and the solid phases, so increasing initial concentrations would enhance the adsorption capacity of arsenic.

3.5. Adsorption Isotherm. Isotherms study can describe how an adsorbate interacts with adsorbent. The isotherm provides a relationship between the concentration of As(V) in solution and the amount of As(V) adsorbed on the solid phase when both phases are in equilibrium. Figures 5, 6, 7, and 8 show the equilibrium isotherms for the adsorption of As(V) onto *S. persica* stem ash, and the equilibrium adsorption data were analyzed by using the Langmuir and Freundlich isotherm models. The Langmuir isotherm model is valid for monolayer adsorption onto surface containing a finite number of identical sorption sites which is presented by the following equation

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}, \quad (3)$$

where q_e is the amount of metal adsorbed per specific amount of adsorbent ($\mu\text{g/g}$), C_e is equilibrium concentration of the solution ($\mu\text{g/L}$), and q_m is the maximum amount of As(V) required to form a monolayer ($\mu\text{g/g}$). The Langmuir equation can be rearranged to a linear form for the convenience of plotting and determining the Langmuir constants (K_L) and the maximum monolayer adsorption capacity of *S. persica* (q_m). The values of q_m and K_L can be determined from the linear plot of $1/q_e$ versus $1/C_e$:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{q_m K_L} \frac{1}{C_e}. \quad (4)$$

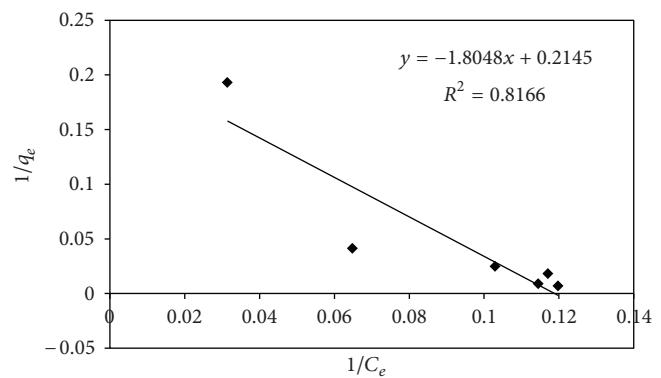


FIGURE 5: The Langmuir isotherm for adsorption of As(V) onto *S. persica* stem ash 300°C.

The Freundlich equation is purely empirical based on sorption on heterogeneous surface, which is commonly described by the following equation:

$$q_e = K_f C_e^{1/n}, \quad (5)$$

where K_f and $1/n$ are the Freundlich constants related to adsorption capacity and adsorption intensity, respectively. The Freundlich equilibrium constants were evaluated from the intercept and the slope, respectively, of the linear plot of $\log q_e$ versus $\log C_e$ based on experimental data. The Freundlich equation can be linearized in a logarithmic form for the determination of the Freundlich constants as shown below:

$$\log q_e = \log K_f + \frac{1}{n} \log C_e. \quad (6)$$

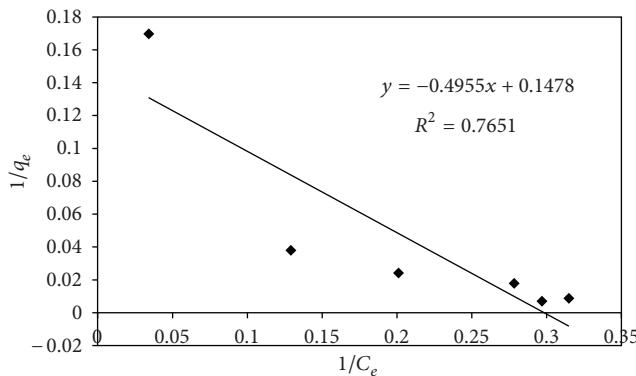
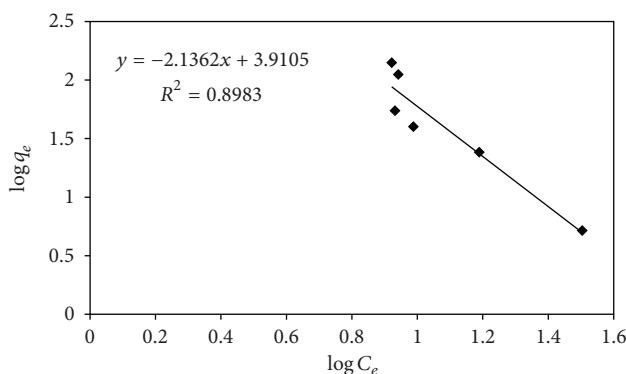
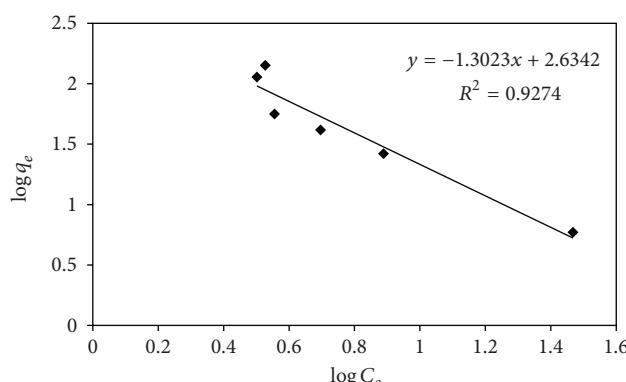
The isotherms based on the experimental data and the parameters obtained from nonlinear regression by both models are shown in Figures 5, 6, 7, and 8. In addition, the calculated parameters of the Langmuir and Freundlich isotherm models for *S. persica* stem ash at 300°C and 500°C, as well as the correlation coefficients (R^2), are listed in Table 2. The Freundlich models showed significantly higher correlations than the Langmuir models. Therefore, the Freundlich adsorption isotherm is suitable for describing the adsorption equilibrium of As(V) onto *S. persica* stem ash. As mentioned above, K_f is the equilibrium constant indicative of adsorption capacity and higher K_f means that the adsorption capacity is higher. Furthermore, the q_m value of ash at 500°C was higher than that of ash at 300°C, which indicates that ash 500°C is a better adsorbent for As(V) removal than ash at 300°C. As noted above, the parameter K_L is a function of the strength of adsorption. The larger K_L means that the adsorption bond is stronger.

4. Conclusions

Arsenic is present in water as a result of both natural and anthropogenic activities. Drinking water poses the greatest threat to public health from arsenic. Arsenic dissolved in water is acutely toxic and can lead to a number of health

TABLE 2: Pertinent parameters of the adsorption isotherm equations at $23 \pm 2^\circ\text{C}$.

Temperature of ash	The Langmuir isotherm			The Freundlich isotherm		
	q_m ($\mu\text{g/g}$)	k_L ($\text{L}/\mu\text{g}$)	R^2	k_f	n	R^2
300°C	4.66	0.12	0.8166	8137.7	0.47	0.8983
500°C	6.77	0.3	0.7651	430.7	0.77	0.9274

FIGURE 6: The Langmuir isotherm for adsorption of As(V) onto *S. persica* stem ash 500°C.FIGURE 7: The Freundlich isotherm for adsorption of As(V) onto *S. persica* stem ash 300°C.FIGURE 8: The Freundlich isotherm for adsorption of As(V) onto *S. persica* stem ash 500°C.

problems. At the present study, the adsorption ability of the *S. persica* stem ash (300°C and 500°C) is evaluated for As(V) as an adsorbate. Batch adsorption tests demonstrate that the adsorption is affected by various conditions such as initial pH, adsorbent dosage, contact time, and initial As(V) concentration. From the present study, it can be seen that *S. persica* stem ash can be used effectively for the removal of As(V) from aqueous solutions. This adsorbent was able to remove up to 99% of As(V) from solutions whose initial concentration varied between 50 and 500 $\mu\text{g/L}$. In addition, final concentration of As(V) reached lower than 10 $\mu\text{g/L}$ as a maximum contaminant level in drinking water. The removal of As(V) from aqueous solutions is induced by adsorption on surface sites of the solid for low As(V) concentration while both adsorption and internal exchange take place for high concentrations.

Acknowledgments

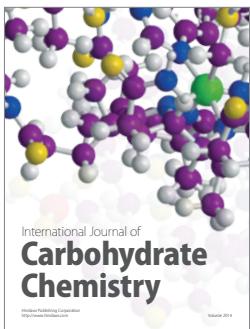
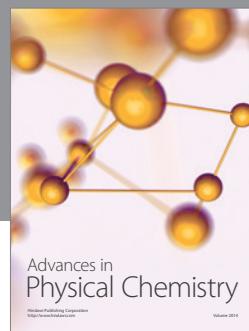
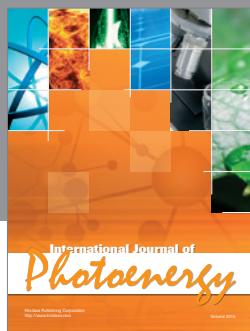
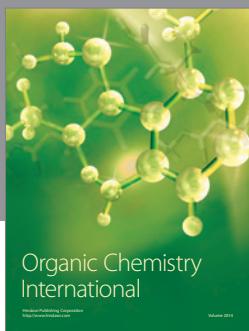
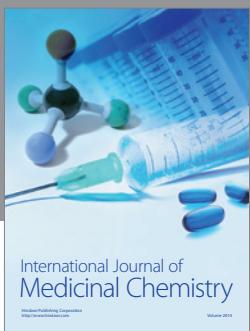
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