

Removal of Arsenic from Water Using Aluminum Nanoparticles Synthesized through Arc Discharge Method

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ABSTRACT: *The present study describes a novel procedure for As (V) removal from water using pure Al nanoparticles (AlNps) prepared by arc discharge technique. Some spectroscopic and microscopic techniques such as X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), Brunauer–Emmett–Teller (BET) and Barrett–Joyner–Halenda (BJH) corroborated the structure of the prepared nanoparticles. From TEM image, the average size of nanoparticles was nearly calculated 15 nm. To confirm the removal of arsenic, Inductive Coupled Plasma Optical Emission Spectroscopy (ICP-OES) was used. The effects of some parameters such as contact time, adsorbent dose, As (V) initial concentration, pH, and ionic strength were investigated. In optimum conditions, for a solution with the initial concentration of 30 mg/L of arsenic (V), the maximum removal (92%) took place after 3h at pH of 3. The adsorption followed the pseudo-first-order kinetic model. The equilibrium data fitted well to Langmuir adsorption isotherm which suggested monolayer adsorption. Adsorption capacity was calculated 55.5 mg/g through Langmuir isotherm which confirmed AlNps present an outstanding ability to remove As (V) not only due to a high surface area and small particle size but also because of their great inherent action.*

KEYWORDS: *Arsenic; Al nanoparticles; Arc discharge; Adsorption; Langmuir isotherm.*

INTRODUCTION

Arsenic is a toxic and carcinogenic metalloid that is introduced in the aqueous system through natural as well as anthropogenic sources. It exists in both organic and inorganic forms in nature. Inorganic arsenic, which is more toxic than organic arsenic, is mostly found in natural water systems and has two different oxidation states, that is, trivalent and pentavalent [1]. The speciation

of arsenic highly depends on solution pH. Pentavalent arsenic (As (V), arsenate) is stable in oxidative conditions, while trivalent arsenic (As (III), arsenite) is stable in reductive conditions [2]. Contamination of potable ground water with arsenic and its removal in water treatment have become the focus of increasing attention worldwide. Elevated arsenic concentrations

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in excess of 10 µg/L (the maximum level recommended for potable waters by the World Health Organization, 1993 [3]) have been found in different parts of the world [4]. Several approaches for metal-treatment waste water have been described including chemical and surface chemistry processes. Sensitive operating conditions, low efficiency, and production of secondary sludge demanding additional expensive disposal are inherent limitations in the application of these methods [5-7]. Various technologies such as precipitation, ion exchange, membrane filtration and reverse osmosis have been applied for the removal of pollutants from waste waters [8-9]. However, these methods have somehow proven disadvantageous in as much as they require expensive equipments and/or continuous need of chemicals [10]. Moreover, sometimes the above-mentioned methods fail to meet the environmental protection agency requirements [11]. Considering limitations of conventional methods for metal removal, the most promising alternative appears to be the adsorption process [12]. Adsorption is a mass transfer process by which a substance is transferred from the liquid phase to the surface of solid, and becomes bound by physical and chemical interactions. Adsorption phenomena are operative in most natural physical, biological and chemical systems. Up to date, a wide range of nanoparticles such as iron oxide nanoparticles [13], nano- Al_2O_3 [14], iron nanoparticles [15] and other nanoparticles [16] have been reported to demonstrate arsenic removal efficiency. Until now, no report has been made on using AlNps for arsenic removal. Here, we report the efficiency of AlNps, fabricated through arc discharge method, for removing arsenic from water. Adsorption capacity of these nanoparticles was excellent than those that have been previously reported.

EXPERIMENTAL SECTION

Materials

All chemicals were of analytical grade and used without further purification. ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ (>99.9%), HCl, NaOH) were obtained from Aldrich Chemical Co. or Merck Co, Germany. Solutions of As were prepared with deionized water using $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$. Either 1 M HCl or NaOH was used for adjustment of pH using a pH meter (Suntex model sp-701).

Preparation of AlNps

AlNps were fabricated through a modified arc discharge technique. Briefly, two 95.5% pure aluminum

rods with a diameter of 2 mm and length of 40 mm were used as electrodes: one as a movable anode and the other as a static cathode. The distance and angle between them was set at 1 mm and 85°, respectively. A current of 50 A/cm² was passed through Ethylene Glycol (EG)-submerged aluminum electrodes until explosions occurred (1–10 ms). The cooled metal vapor in ethylene glycol formed the primary particles needed for nucleation mechanism, giving AlNps dispersed in ethylene glycol.

Adsorption experiments

Tests were conducted at atmospheric pressure in 50 mL polyethylene bottles, in which the synthesized AlNps and selected concentrations of As (V) solution (0, 5, 10, 15, 20, 25 and 30 mg/L) were placed. Ionic strength of the solutions was adjusted by NaCl to 0.001, and 0.01 mol/L. The pH was set at 3, 5, 7 and 9. Each bottle rotated at 400 rpm for 3h. After filtration through the Whatman filter paper, the concentration of arsenic was measured by an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES).

The amount of adsorbed As (V) per gram of AlNps at equilibrium, q_e (mg/g), and the removal percentage, (% removal), was calculated by equations given below:

$$q_e = (C_0 - C_e) / W \quad (1)$$

$$\% \text{ Removal} = (C_0 - C_e) / C_0 \times 100 \quad (2)$$

Where C_0 and C_e are the initial and equilibrium concentrations of As (V) in mg/L, respectively. V is the volume of As (V) solution (L) and W is the weight of adsorbent (g) [17].

Characterization

X-ray diffractometer (XRD, Philips X'pert 1710) with $\text{Co-K}\alpha$ ($\lambda = 1.78897 \text{ \AA}$) irradiation was used for the structural characterization of the synthesized nanoparticles. Scanning electron microscopy (SEM, Philips XL 30 and S-4160 instruments) and Transmission electron microscopy (TEM, Zeiss-EM10C) images were used for the evaluation of nanoparticles morphology. Brunauer–Emmett–Teller (BET, PHS-1020) analysis was used for determination of the pore volume and average pore size of nanoparticles. Inductive Coupled Plasma Optical Emission Spectroscopy (ICP-OES, VISTA-PRO) was used To confirm the removal of arsenic.

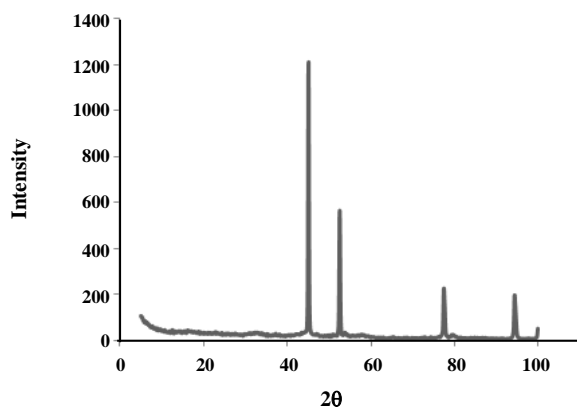


Fig. 1: XRD pattern of the synthesized AlNps.

RESULTS AND DISCUSSION

X-Ray Diffraction (XRD) pattern of the synthesized AlNps is shown in Fig. 1. As seen in this Figure, the XRD pattern exhibits diffraction peaks at $2\theta = 45.0^\circ$, 52.4° , 77.4° , and 94.3° which corresponds to the (111), (200), (220), and (311) crystallographic planes. The observed diffraction peaks agree well with the body centered cubic (bcc) structure of AlNps.

Nanoparticles morphology was evaluated by SEM (Fig. 2a) and TEM (Fig. 2b and c) images. Spherical in shape with a smooth surface morphology of the nanoparticles are clearly seen in these images. A typical way to show the particle size and its distribution is in the form of a number-frequency histogram. The obtained histogram (Fig. 2d) confirmed the narrow size distribution for 30 observed nanoparticles of Al with a 15 nm average value.

The porous nature of nanoparticles was clearly proved by the BJH method which showed the radii of pores to be about 3.7 nm. The increased active site of the adsorbent is studied by the nitrogen adsorption-desorption isotherms from the BET analysis and a type-IV hysteresis loop is detected. The specific surface area and the total pore volume of the mesoporous adsorbent calculated as $42 \text{ m}^2/\text{g}$ and $0.16 \text{ cm}^3/\text{g}$, respectively (Fig. 3).

Effect of AlNps dosage and initial As (V) concentration

Fortunately, 100% removal of arsenic *via* AlNps (0.8 g/L) was achieved in 3h from solutions containing 5 mg/L of As (V) at pH = 3 (Fig. 4). Gradual increase of As (V) concentration from 5 mg/L to 30 mg/L slightly decreased percent arsenic removal to 92%. Furthermore,

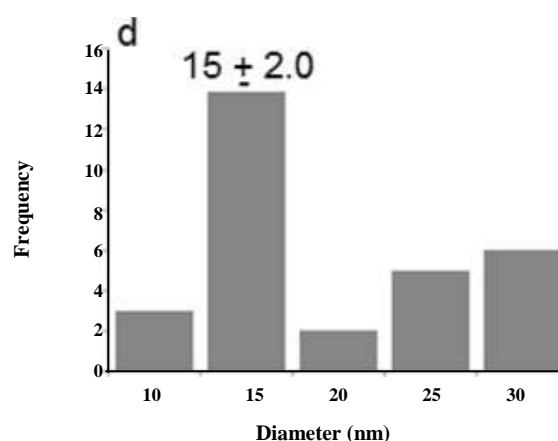
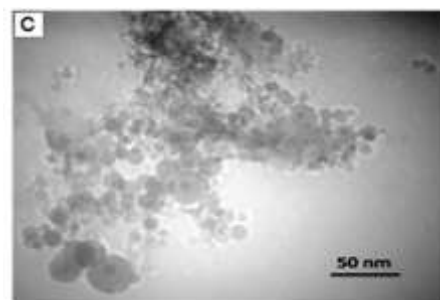
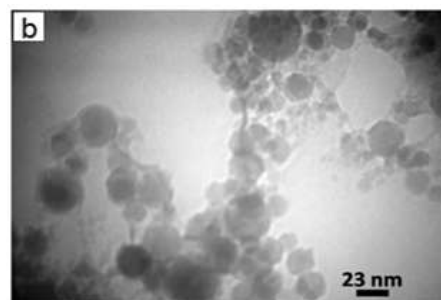
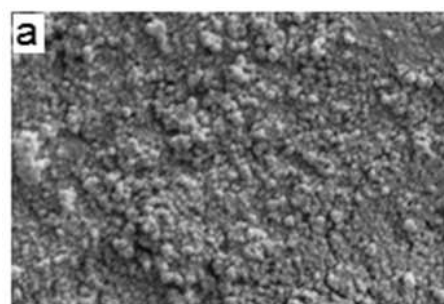


Fig 2: SEM (a), TEM (b and c) images of AlNps and histogram generated from the sizes of nanoparticles using the SEM images (2d).

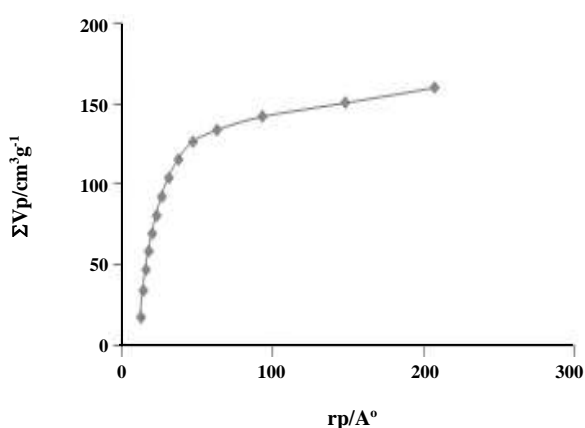
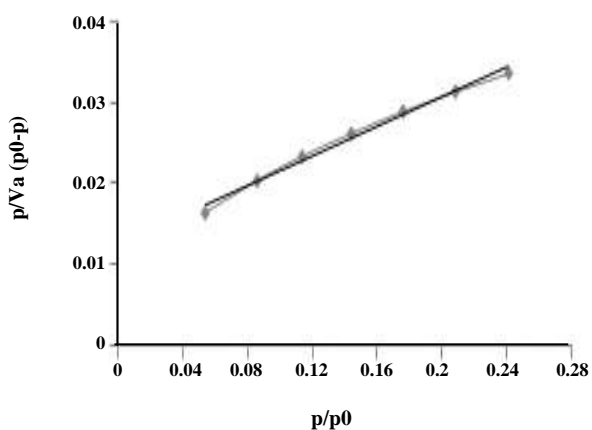
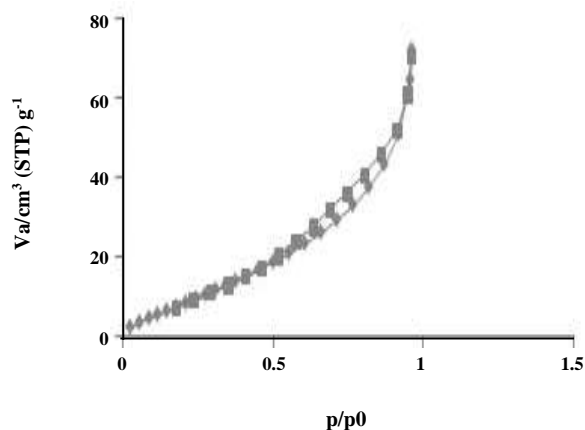


Fig. 3: N_2 adsorption-desorption analysis, Brunauer-Emmett-Teller (BET) analysis and Barrett-Joyner-Halenda (BJH) analysis of the synthesized AlNps.

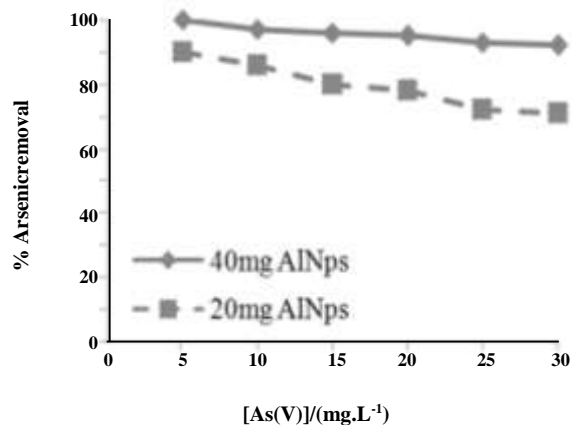


Fig. 4: Removal of arsenic from water as a function of initial As (V) concentration for different AlNps dosage, at 0.01M NaCl ionic strength and pH = 3, during 3h.

decreasing the concentration of the adsorbent from 0.8 g/L to 0.4 g/L was resulted in arsenic removal from 100% to 90% for the As (V) solutions with initial concentration of 5 mg/L. This decrease was slightly more for solutions containing higher initial concentration of As (V). Due to decreasing the adsorbent amount from 0.8 g/L to 0.4 g/L for a solution with initial concentration of 30 mg/L As (V), the arsenic removal went down by 71%. Actually, with increasing initial arsenic concentration, competition for the active adsorption sites rises and the adsorption process will increasingly slowed down.

Effect of pH

Since the surface charge of an adsorbent can be modified by changing pH of the solution, pH is one of the most important parameters affecting the adsorption process. Here, four different pH values were investigated. Particularly, 40 mg of adsorbent (AlNps) was added to an arsenic solution (50 ml, 5-30 mg/L), in a 60 mL polyethylene bottle, and the pH was adjusted to 3 via 1 M NaOH or HCl. Similarly, three other samples were made at pH 5, 7 and 9. With increasing pH from 3 to 9, the corresponding adsorption dropped from 92% to 56% (Fig. 5). The reason for this trend is hastened corrosion of Al⁰ in acid conditions. At lower pH, in fact acidic environment, intensify the speed of oxidation and a layer of hydroxide covers the external surface of Al Nps which ends up removing of arsenate ions with physical adsorbing on aluminum hydroxide [18].

Effect of reaction time

In order to establish equilibration time for maximum removal, the adsorption of As (V) on AlNps was studied as a function of contact time (Fig. 6). The removal efficiency was directly proportional with the contact time and increased with the elapsed period. For 30 mg/L initial As (V) concentration and AlNps dosage of 0.8 g/L, at pH = 3, arsenic removal of 92% was found after 3h, which remained stable over the time. So, the time required for equilibrium adsorption was 3h. The higher As (V) removal with elapsing time is due to the superior surface area of our AlNps.

Effects of ionic strength

Two sets of solutions with differing ionic strength (0.01 and 0.001M NaCl) were prepared by adding NaCl to the samples containing 5-30 mg/L As (V) at pH = 3. The effect of ionic strength on the removal of As (V) in 3h was measured by adding identical quantity of AlNps adsorbent (0.8g/L) to these 12 samples (Fig. 7). Increasing the ionic strength of solutions had a direct correlation with the arsenic removal. In other words the efficiency of As (V) removal increased in the presence of Cl⁻.

Adsorption isotherms

The adsorption processes of As (V) on AlNps adsorbent were tested with two commonly used experiential adsorption models, Freundlich and Langmuir, which correspond to heterogeneous and homogeneous adsorbent surfaces. The linear form of Langmuir equation is as follows [19]:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{K_L q_m C_e} \tag{3}$$

The equilibrium concentration (C_e) is given in mg/L. The maximum adsorption capacity (q_m) is in mg/g. Finally, the Langmuir constant (K_L), which represents the affinity between the solute and the adsorbent, is given in terms of L/mg. Values of K_L and q_m were calculated from the slope and intercept of the linear plot of 1/q_e vs. 1/C_e (Fig. 8a). The maximum adsorption capacity attained for AlNps was 55.5 mg/g that based on our research, this was a very ideal and salient amount than those previously reported for another nanoparticles. The essential characteristics of the Langmuir isotherm can be expressed by separation or equilibrium parameter, a dimensionless constant asper correlation [17]:

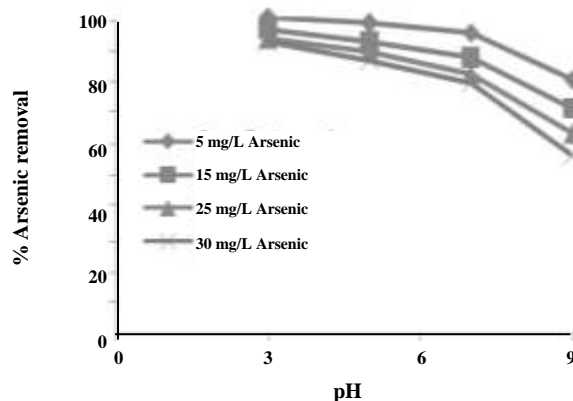


Fig. 5: Removal of As (V) from water as a function of pH for 0.8 g/L of AlNps solutions containing different concentrations of As (V), at 0.01M NaCl ionic strength and during 3h.

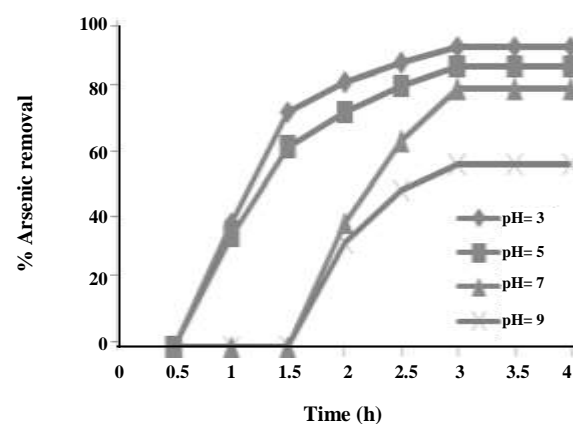


Fig. 6: Adsorption of As (V) on AlNps as a function of time, using 30 mg/L As (V), 0.8 g/L AlNps, 0.01M NaCl solution, at different pH.

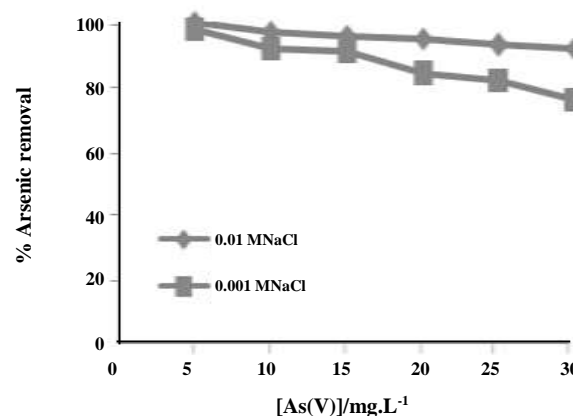
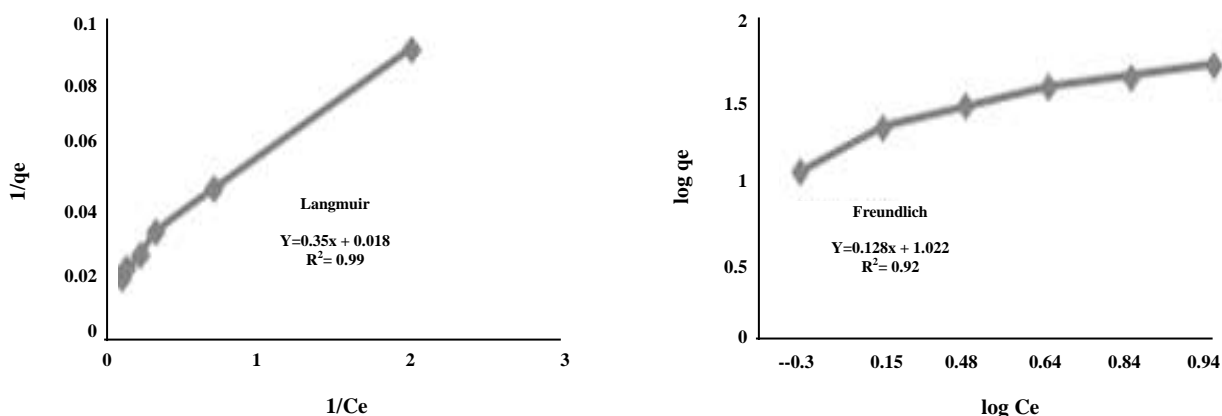


Fig. 7: Adsorption of As (V) on AlNps as a function of ionic strength and initial As (V) concentration. Reaction conditions: pH = 3, initial Al Nps weight = 0.8 g/L and reaction time = 3h.

Table 1: Comparison of adsorption capacity for removal of arsenic (V) with various nanoadsorbents.

Adsorbent	Adsorption capacity (mg/g)	pH	Concentration range(mg/L)	Ref.
Al nanoparticles	55.5	3	5-30	Present work
Fe ₂ O ₃	4.6	7	1-4	16a
Fe ₃ O ₄	0.2	9	0.1-2	16b
Fe ₃ O ₄ -γFe ₂ O ₃	4.85	6.5	2	16c
Crystalline TiO ₂	37.5	7	-	16d
Fe ₃ O ₄ – BNNTs	32.175	6.9	1-40	16e
NZVI–RGO	29.04	7	1–15	16f
Magnetite nanoparticles	3.7	2	2	16g
NZVI–RGO nanoparticles	29.04	7	1-10	16h
Al ₂ O ₃	0.17	6	200–600	16i
Al	40.9	6.5	5-30	16j

**Fig. 8: Langmuir isotherm (a) and Freundlich isotherm (b) at pH = 3. The concentration range of As (V) is 5–30 mg/L while the adsorbent concentration is 0.8 g/L.**

$$R_L = 1 / (1 + K_L C_o) \quad (4)$$

R_L describes the nature of the adsorption process as follows:

- $R_L = 0$, Irreversible
- $R_L = 1$, Linear
- $R_L > 1$, Unfavorable
- $0 < R_L < 1$, Favorable

The values of R_L in the range of 0–1 indicates that the adsorption process is favorable for adsorbent. R_L value for this study was found 0.06, suggesting very favorable adsorption of As (V) onto the AlNps. The adsorption capacity of AlNps was compared with other previously reported adsorbents in Table 1. As can be seen in this table, AlNps,

produced during this study, have shown a magnificent and efficient function in removing poisonous arsenic.

The Freundlich isotherm speculates the heterogeneity of the surface and that the adsorption occurs at sites with different energy of adsorption. The energy of adsorption is a function of the surface coverage. Linearized Freundlich isotherm model as shown in Equation (5) was used to determine the values of Freundlich constants from slope and intercepts [19].

$$\log(q_e) = \log(K_f) + (1/n_f) \log(C_e) \quad (5)$$

where, K_f (mg/g) and n are the Freundlich parameters related to the adsorption capacity and adsorption intensity, respectively. They can be obtained from

Table 2: Langmuir and Freundlich adsorption isotherm parameters for As (V) adsorption on Al Nps at pH = 3.

Langmuir model			Freundlich model		
R ²	K _L (L/mg)	R _L	R ²	K _F (mg/g)	N
0.99	0.52	0.06	0.92	10.5	7.8

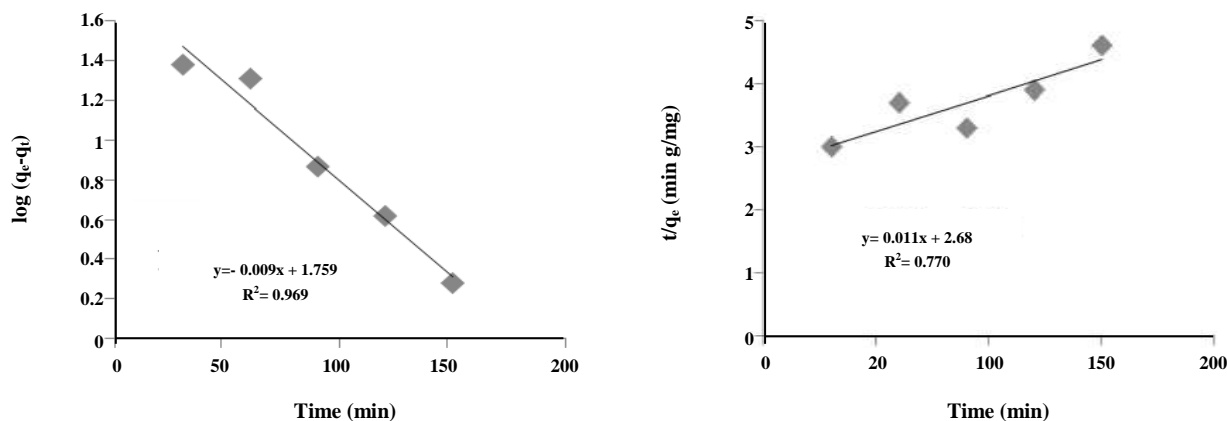


Fig. 9: The pseudo first-order plot (a) and pseudo second-order plot (b).

the intercept and slope of the plot of $\log q_e$ versus $\log C_e$, correspondingly (Fig. 8b). Freundlich parameters (K_F and n) indicate whether the nature of adsorption is either favorable or unfavorable. Values $1 < n < 10$, are considered favorable [19]. The Freundlich parameter for n is 7.8 indicating that arsenic favorably adsorbs on the adsorbent. The results of adsorption isotherms are listed in Table 2. Analysis of data for selection of the best fit isotherm was carried out by linear regression analyses of these models and comparison of the correlation coefficients (R^2) [17]. The adsorption data fits the Langmuir model ($R^2 = 0.99$) better than Freundlich model ($R^2 = 0.92$) that suggests a monolayer adsorption on the AlNps (Table 2).

Adsorption kinetics

In order to suggest the mechanism of adsorption, kinetic models such as first-order, and pseudo-second-order were used. The first-order rate Lagergren model is as follows [20]:

$$\log(q_e - q_t) = \log q_e - \frac{K_1}{2.303} t \tag{6}$$

Where q_t is the amount of adsorbed As (V) on the adsorbent at time t and K_1 is the rate constant of the pseudo-first-order (min^{-1}). K_1 and q_e can be calculated

respectively from the slope and intercept of the plot $\log (q_e - q_t)$ versus t (Fig. 9a). It is found from Table 3 that the q_e value obtained by pseudo-first-order kinetic model ($q_{\text{cal.}}$) is in good agreement with the measured experimentally ($q_{\text{exp.}}$), suggesting the adsorption is first-order reaction. The value of correlation coefficient (R^2) is 0.969, indicating also that the adsorption is the pseudo-first-order reaction.

Experimental data were also tested by the pseudo-second-order kinetic model which is given in the following form [21]:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \tag{7}$$

Where K_2 is the rate constant of the equation ($\text{g/mg}\cdot\text{min}$). q_e ($q_{\text{cal.}}$) and K_2 can be calculated respectively from the slope and the intercept of the plot t/q_e versus t (Fig. 9b). R^2 for pseudo -second-order is equal to 0.770 and the $q_{\text{cal.}}$ is not agree with $q_{\text{exp.}}$ (Table 3). Both parameters suggest that the adsorption of As (V) has not followed the pseudo second-order kinetic model.

In view of these results, it can be said that the pseudo-first-order kinetic model provided a good correlation for the adsorption of As (V) onto AlNps in contrast to the pseudo-second-order model.

Table 3: Kinetic parameters obtained from pseudo-first-order and pseudo-second-order for As (V) and adsorption onto AlNps (pH = 3, adsorbent concentration: 0.8 g/l, volume of solution: 50 mL, initial concentration = 30 mg/L).

Pseudo-first-order				Pseudo-second-order			
q _{exp.} (mg/g)	q _{cal.} (mg/g)	K ₁ (l/min)	R ²	R ²	K ₂ (g/mg-min)	q _{cal.} (mg/g)	q _{exp.} (mg/g)
34.5	57.4	0.0207	0.969	0.770	0.000313	90.9	34.5

CONCLUSIONS

This study was conducted to investigate performance of arsenic removal by AlNps fabricated through a modified arc discharge method under different conditions. It was found that these nanoparticles were efficient reagent with high arsenic removal capacity. Arsenic was removed continuously as the contact time, adsorbent concentration and ionic strength were separately increased through experiments. However, removal of the toxic arsenic was decreased by increasing pH and arsenic initial concentration in the solution. Optimum (92%) removal of arsenic was achieved via AlNps (0.8g/L) in 3h from solutions containing 30 mg/L of As (V), at pH = 3. Also, It was obtained (79%) at pH = 7 with the same situations. The maximum adsorption capacity of nanoscale aluminum particles determined by the Langmuir equation was an exceptional 55.5 mg/g, which shows they could be an excellent candidate to help with removal of arsenic contamination from aqueous solutions.

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