

Removal of Arsenic From Aqueous Solution Using Silica Ceramic: Adsorption Kinetics and Equilibrium Studies

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ABSTRACT: The removal of As(III) ions from aqueous solutions has been investigated using silica ceramic under different experimental conditions of initial arsenite concentration, adsorbent dose, temperature and pH. Better removal was obtained at temperature 25°C and pH 7.5. The adsorption data have been explained in terms of Langmuir and Freundlich equations. The sorption kinetics was tested for the pseudo-first order, pseudo-second order reaction and intra-particle diffusion, at different experimental conditions. The results revealed that the adsorptions of As(III) onto silica ceramic, which were found to well, fit by the Freundlich isotherm. The rate constants of sorption for kinetic models were calculated and good correlation coefficients ($R^2 > 0.9974$) obtained for the pseudo-second order kinetic model. The result indicates that under the optimum conditions, the maximum adsorption value (1.7886 mg/g) comply that the silica ceramic is an effective adsorbent for arsenite.

Key words: Arsenic, Adsorption, Isotherms, Silica ceramic, Kinetics

INTRODUCTION

Arsenic is one of the toxic contaminants found in natural and industrial wastewaters. Groundwater arsenic contamination is serious threat in Bangladesh and West Bengal (India) because groundwater is the main source of drinking water (Dixit and Hering, 2003; Pal *et al.*, 2002). In recent years, arsenic contamination of water and groundwater about the effect of toxicity has become an increasing concern on a global perspective. Long-term exposures to arsenic levels can result in permanent and severe damage to human health. Arsenic toxicity causes various health hazards, such as skin lesions, liver, lung, kidney, bladder, and nerve tissue injuries (Roberts *et al.*, 2004; Thirunavukkarasu *et al.*, 2003). Arsenic exposure leads to black foot disease, diffused and spotted melanosis, diffused and spotted keratosis, nonpitting oedema, and gangrene (Biswas *et al.*, 1998).

Various methods of arsenic removal from contaminated water have been applied including

chemical precipitation, membrane process, ion exchange and adsorption. Adsorption is widely used to remove heavy metals from aqueous solutions as the most effective method using low-cost adsorbents (Aultundogan *et al.*, 2000). Several studies have demonstrated that arsenic removal can be achieved by technologies such as, activated alumina sorption, polymeric anion exchange sorption by iron oxide-coated sand (Zang and Itoh, 2005; Gupta *et al.*, 2005; Zeng, 2003), iron oxide-coated cement (Kundu and Gupta, 2007), coagulation with ferric chloride (Meng *et al.*, 2000), pressurized granulated iron particles, iron oxide-doped alginate manganese dioxide coated sand, polymeric ligand exchange, zeolite (Xu *et al.*, 2002), Portland cement (Kundu *et al.*, 2004), activated red mud (Genc *et al.*, 2003; Aultundogan, *et al.*, 2002), Activated Alumina (Singh and Pant, 2004) and zero valent iron, and these have been tried in the laboratory and/or in the field. Present study carried out to evaluate the performance of silica ceramic for As(III)

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removal. The main objectives of this study were to investigate the adsorption characteristics of silica ceramic for the removal of arsenite from aqueous solution. The effects of process parameters such as the effect of initial As(III) concentration, dose, temperature, pH on the adsorption process were investigated. Kinetics and equilibrium experiments have been performed under diverse conditions.

MATERIALS & METHODS

In this study silica ceramic have been used as an adsorbent (size $0.42 \pm 0.02 \text{ mm}$) obtained from EKOAIRANDO Co. Ltd., Kochi, Japan. This silica ceramic was made from special volcano ash named Akadama mud and burned at 400°C up to 20 minute in controlled conditions, which is commercially available in the market. The raw material collected from Kanuma City at Tochigi prefecture in Japan. The physical and chemical properties of silica ceramic are shown in (Table 1). All reagents were of analytical grade, which were purchased from Nacalai Tesque, Inc. and Kanto Chemical Co., Inc. Japan. A synthetic stock solution arsenite was made using standard (1000 mg/L) As(III) solutions and deionized water without further purification. Standard acid and base solutions (0.1N HCl and 0.1N NaOH) were used for pH adjustments. Further working solution were freshly prepared from stock solution for each experimental run. Arsenic concentration determination were carried out using a more sensitive and sophisticated instrument, atomic absorption spectrometer with flow injection analysis system (AAAnalyst 200, Perkin-Elmer, Singapore) by using EDL at a wavelength 193.7 nm in order to get a more accurate measurement. The surface area of the silica ceramic was measured by N_2 adsorption using single point Brunauer, Element and Teller (BET) (Micrometric ASAP 2020, US) procedure.

The effect of initial pH (4.0, 7.5 and 10.7) on arsenic uptake, experiments were performed with initial As(III) concentrations 50 ml of 2 mg/L, adsorbent dose of 20 g/L, and incubation temperature at 25°C . The effect of the adsorbent dosages were investigated using 10-100 g/L of adsorbent at a fixed pH of 7.5 and 25 ml of 0.5 mg/L As(III) solution in 50ml flasks. These flasks were shaken for 3h to reach the shake equilibrium at 25°C of fixed temperature. Twenty-five mL of

different known initial As(III) concentrations 20, 39, 60, 76, 98 mg/L were used to examine the effect of Initial concentration and contact time. Where adsorbent dose were used 80 g/L at pH 7.5 and 25°C temperature. Samples of the ceramics adsorbent were placed in labeled round-bottomed in 50 ml flasks. The sample were collected by predetermine time interval. Effect of temperatures (15°C , 25°C , 35°C , and 45°C) were studied with an initial As(III) concentration of 2 mg/L of 50 ml solution and adsorbent dosage of 20 g/L at pH 7.5. Adsorption isotherm studies were conducted with 25 ml of varying initial As(III) concentrations (20, 39, 60, 76 and 98 mg/L), fixed adsorbent dose of 80 g/L at pH 7.5 and 25°C . Each experiment the flasks were sealed with airtight cap and samples were shaken on the mechanical shaker at 150 rpm. Optimizations of parameters adsorbent dosage, contact time, pH, and various initial concentrations, temperature were optimized for maximum adsorption by the method of continuous variation following batch adsorption experiments. The quantity of adsorbed arsenic was calculated by the difference of the initial and residual amounts of arsenic in solution divided by the weight of the adsorbent by the following equation:

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

The amount of adsorption at time t , q_t (mg/g), was calculated:

$$q_t = \frac{(C_0 - C_t)V}{M} \quad (2)$$

Where, q_e (mg/g) is the amount of As(III) anion adsorbed per gram of the ceramic, C_0 (mg/L) the initial concentration of the solution of As(III), C_t (mg/L) is the concentration at any time t , C_e (mg/L) the equilibrium concentration of the solution of As(III). V (L) is the volume of the solution and M (g) is the mass of the adsorbent. The percentage of As(III) removed by the silica ceramic shows the efficiency of removal. It is determined from the ratio of concentration of As(III) present in the solution and particulate phases using the equation:

$$R_e = \frac{C_0 - C_e}{C_0} \times 100 \quad (3)$$

Where R_e is the percentage (%) of As(III) adsorbed, C_0 and C_e are the initial and equilibrium concentration of As(III) respectively.

RESULTS & DISCUSSION

Scanning electron microscopy using JEOL-JSM-6500F was performed on silica ceramic at different magnification to determine surface structure. The working tension was 15 KV. Scanning electron micrographs for intra structure of silica ceramic is shown in Fig.1 (a) and (b). The figure indicates that silica ceramic can be used as a potential adsorbent. The chemical composition of silica ceramic (Table 1) showed that it was mainly composed silica as SiO_2 (53.97%), Aluminum as Al_2O_3 (40.77%), iron as FeO (2.69%), and sodium as Na_2O (2.57%).

Table 1. Physical and Chemical Properties of Silica Ceramic

Physical properties		Chemical compositions	
Parameters	kind/Value	Parameters	Value (%)
Color	Red-brown	SiO2	53.97
Size (Spherical mm)	0.42±0.02	Al2O3	40.77
Particle form	Sphere	FeO	2.69
Bulk density (g/cm3)	0.808	Na2O	2.57
Porosity (%)	69.5		
Pore volume (cm3/g)	0.2616		
Surface area (m2/g)	211.657		
Pore diameter (A)	49.446		

Effect of pH on the adsorption of As(III) onto silica ceramic was studied at temperature 25°C, initial concentration of 2 mg/L, adsorbent dose 20 g/L, 150 agitation speed are presented in Fig. 2. In this experiment, the duration of arsenic adsorption was kept 60h. The pH range of solution was adjusted between 4 and 10.7. Adsorption was very high in pH range of 6 to 7.5 and maximum adsorption of 0.103 mg/g achieved at pH 7.5. Adsorption value decreases at both lower and higher pH values. The (Fig. 2). shows that silica ceramic adsorbs As(III) better in basic medium. That means favorable adsorption taken place at pH 7.5 (Altundogan *et al.*, 2002) and therefore all further experiments were carried out at pH of 7.5. Similar result were reported for the adsorption of As(III) from aqueous solutions onto activated alumina (Singh and Pant, 2004) and iron oxide-coated sand (Gupta, *et al.*, 2005; Thirunavukkarasu *et al.*, 2003).

The effect of adsorbent dose was studied at a fixed initial concentration of 0.5 mg/L by silica ceramic mass varied from 10 to 100 g/L at solution pH 7.5. The general trend indicates that 10 to 50 g/L of sorbent dose uptake large amount of As(III), rapidly and 60 to 100 g/L dose uptake little amount but slowly (Fig. 3). At 50 g/L As(III), removal efficiency of 82.3% was observed. The highest removal efficiency of silica ceramic for 90 g/L of As(III) was found 96% and thereafter remains nearly constant. The increase in the efficiency of removal may be attributed to the fact that with an increase in the adsorbent dosage, more adsorbent surface is available for the solute to be adsorbed (Genc, *et al.*, 2003).

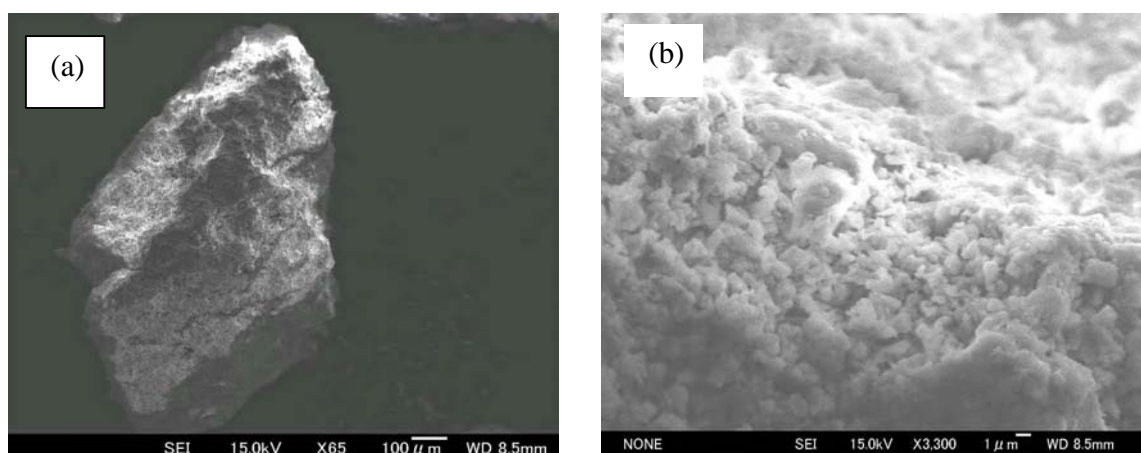


Fig. 1. SEM Photograph of silica ceramic: (a) general intra structure (× 65, 100μm) and (b) intra structure (× 3500, 1μm)

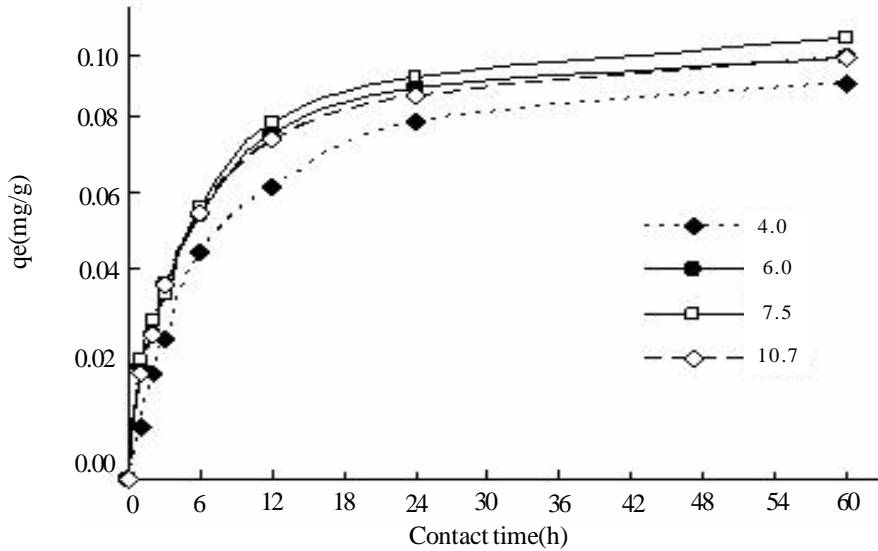


Fig. 2. Effect of pH on adsorption of As(III) by silica ceramic (adsorbent dose 20 g/L, temperature 25°C, As(III) concentration 2 mg/L, contact time 60 h)

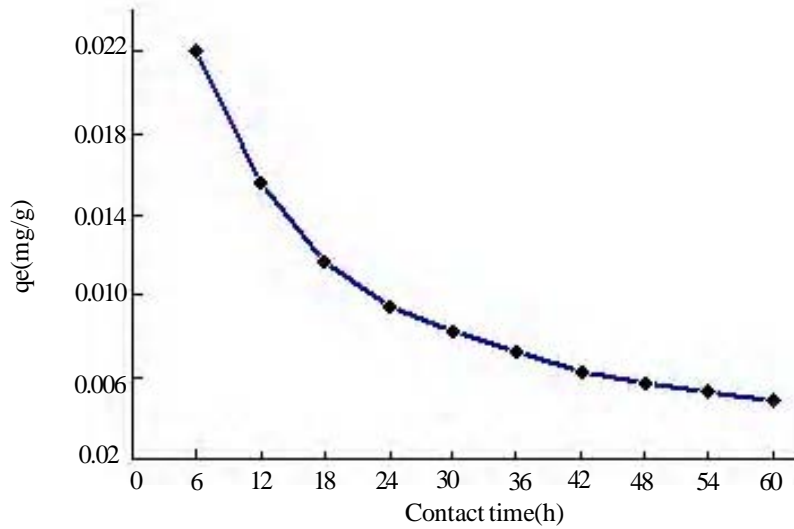


Fig. 3. Effect of adsorbent dose on adsorption of As(III) by silica ceramic (pH 7.5, temperature 25°C, As(III) concentration 0.5 mg/L, contact time 3 h)

A series of experiments were undertaken to study the effect of varying the initial As(III) concentration on arsenic removal kinetics from the solution. (Fig. 4). showed that adsorption of As(III) by silica ceramic increases as the initial As(III) concentration increased. The contact time required to reach the equilibrium of As(III) solution with initial concentration of 20 mg/L was less than 12h, 39 mg/L was 24h, 60 mg/L was 36h, and 76- 98 mg/L was 48h. However, the experimental data were measured at 60h to confirm the complete equilibrium was reached. The study on kinetics of adsorption is quite significant in wastewater

treatment as it describes the solute uptake rate, which in turn controls the residence time of adsorbate uptake at the solid solution interface. It is also an important characteristic in evaluating the efficiency of adsorption. Therefore, three kinetics models were applied mainly, pseudo-first order, pseudo-second order and, intra-particle diffusion model rate equations. Lagergren suggests the pseudo-first order kinetics rate equation (Lagergren, 1898), which is expressed as follows:

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (4)$$

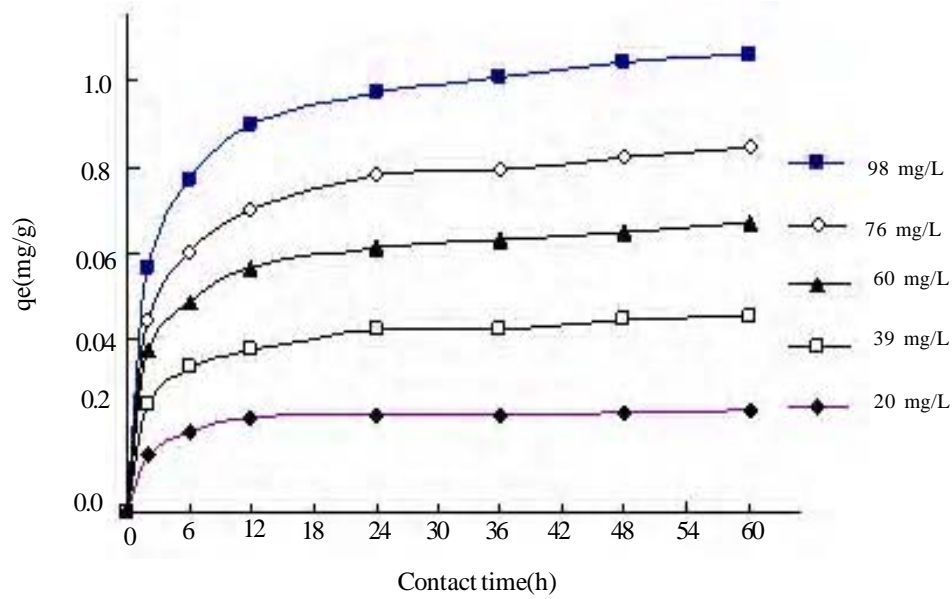


Fig. 4. Effect of initial As(III) concentration by silica ceramic (pH 7.5, adsorbent dose 80 g/L and temperature 25°C)

where k_1 is the pseudo-first order rate constant, q_e represents adsorption capacity. The integrating rate law by applying the initial condition of $t = 0$ to t and $q_t = 0$ to q_t , Eq.(4) becomes:

$$\log(q_e - q_t) = \log q_e - \left(\frac{k_1}{2.303}\right)t \quad (5)$$

Where, q_e and q_t both (mg/g) are the amount of As(III) adsorbed per unit of mass of silica ceramic at equilibrium and time t , respectively, and K_1 the rate constant (1/min). The value for the K_1 was calculated from the slope of the linear plot of $\log (q_e - q_t)$ versus t (Figure not shown). The K_1 values and correlation coefficients R^2 are given in (Table 2). Ho et al. (Ho, *et al.*, 1996) used a pseudo-second order reaction rate equation to study the kinetics of adsorption of heavy metals on peat. The pseudo-second order kinetic model was also applied to assess the kinetics of adsorption of As(III) on silica ceramic. The equation is as follows:

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (6)$$

The integrating rate law, after applying the initial condition of $t=0$ to t and $q=0$ to q , and rearranging it gives as linearized form of pseudo-second order rate kinetics expressed as follows:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (7)$$

In addition, initial rate of adsorption is h is:

$$h = k_2 q_e^2 \quad (8)$$

Where, q_e and q_t both (mg/g) are the amount of As(III) adsorbed per unit of mass of silica ceramic at equilibrium and time t , respectively, and K_2 is the rate constant of pseudo second order adsorption (g/mg min). The kinetics plots between t/q_t versus t were plotted for the different initial concentrations (Fig. 5). Slope and intercept values were solved to give the value of pseudo-second order rate constant (Table 2).

Fig. 3 and Table 2 shows that highly significant regression line ($R^2 > 0.999$) and the data were well fitted only to the pseudo second-order rate equation. The straight line was obtained indicating

Table 2. Pseudo-first order and Pseudo-second order rate constant at 25°C and different initial concentrations

C_0 (mg/L)	q_{exp}	Pseudo-second order model				Pseudo-first order model	
		q_e	$k_2 \times 10^{-3}$	$h \times 10^{-3}$	R^2	$K_1 \times 10^{-3}$	R^2
20	0.241	0.246	33.485	2.024	0.9996	128.738	0.8686
39	0.452	0.465	14.407	3.119	0.9996	170.192	0.952
60	0.673	0.691	9.445	4.503	0.9994	187.925	0.8712
76	0.861	0.885	6.277	4.912	0.999	131.041	0.9324
98	1.069	1.102	5.639	6.846	0.9996	149.234	0.9552

q_e : mg/g, h : mg/g min, k_1 : 1/min, k_2 : g/mg min

that the process follow a pseudo second order kinetics for various concentration of As(III). While the initial As(III) concentration increases from 20 mg/L to 98 mg/L, the adsorption capacity, q_{exp} , increase from 0.241 to 1.069 mg/g. This signifies that the initial As(III) concentration plays a key role in determining the adsorption capacity of As(III) on silica ceramic. It is also observed in Table 2 that when initial As(III) concentration increase from 20 mg/L to 98 mg/L, the rate constant, k_2 decrease from 33.486×10^{-3} to 5.639×10^{-3} g/mg min and values of initial sorption rate h are also increase from 2.024×10^{-3} to 6.846×10^{-3} mg/g min. Above two kinetic equations cannot identify the diffusion mechanisms during the sorption process. Therefore, the experimental

kinetic data were tested against intra-particle diffusion model. The initial rate of intra-particle diffusion can be determined by following equation (Weber and Morris, 1963):

$$q_t = k_i t^{1/2} + C \quad (9)$$

Where, q is the amount of As(III) adsorbed (mg/g) at time t (min), k_i the intra-particle diffusion constant (mg/g min), and C is the intercept. The values of k_i indicate an enhancement in the rate of adsorption. (Table. 3). showed that the increases of initial concentration the k_i , and C also increases. To determine the effect of temperature, on removal of As(III) were performed at four different temperatures, i.e., 15, 25, 35, and 45°C.

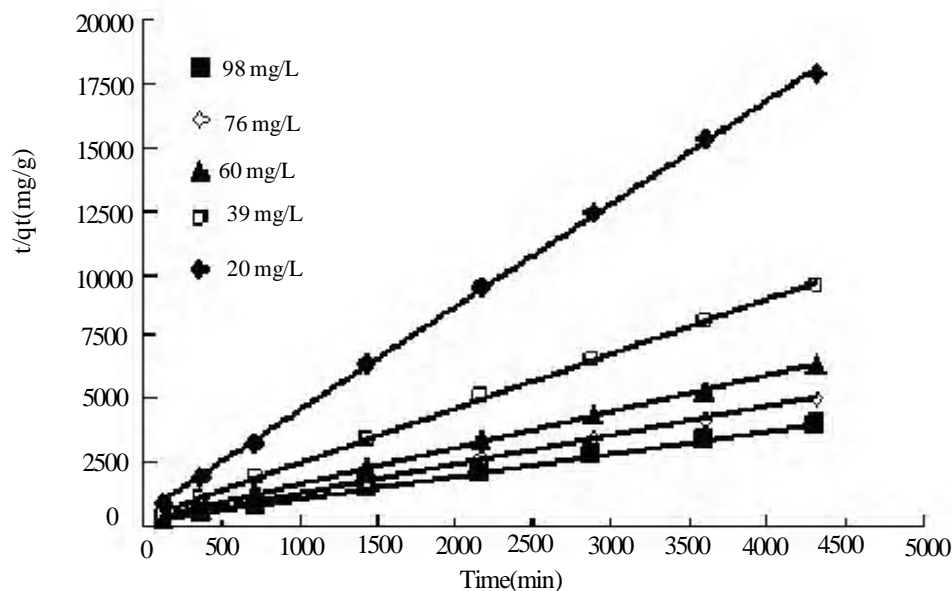


Fig. 5. Pseudo-second order kinetics for adsorption of As(III) onto silica ceramic at different initial As(III) concentration (pH 7.5, adsorbent dose 80 g/L and temperature 25°C)

The results are shown in Fig. 6, which indicates that the As(III) uptake increases from 0.069 to 0.089 mg/g when temperature of the solution increases from 15 to 25°C. After that, sorption uptake decreases slowly from 0.089 to 0.072 mg/g when temperature of the solution increases 25 to 45°C. The adsorption of arsenic on silica ceramic is highest at 25°C. That means the sorption of As(III) is slightly in favor of temperature indicates that the mobility of the arsenic molecule increases with an increase in the temperature.

Fig.7. shows that the linear regression results fitted according to pseudo-second order model. By comparing the coefficient of determination, R², in Table 4 and Fig. 7, it is observed that the pseudo second order model fits the experimental data with higher R² value (0.9974-0.9996) than Lagergren's

pseudo-first order model (0.9881-0.9967). The higher The R² value of pseudo second order model is above 0.9996. Ho and McKay, 1999 for the sorption of As(III) onto zeolite, have reported similar results. The equilibrium adsorption isotherm is of importance in the design of adsorption

Table 3. Intra-particle diffusion rate constant for the adsorption of As(III) onto silica ceramic at different initial concentrations

C ₀ (mg/L)	k _i × 10 ⁻³	C
20	2.8	0.0877
39	5.5	0.152
60	8.2	0.2269
76	10.7	0.2698
98	132	0.3481

C₀: (mg/L), k_i : mg/g min^{1/2}, C: intercept

Table 4. Lagergren Pseudo-first order and Pseudo-second order rate constants, initial As(III) concentration 2mg/L and different temperatures

Temp °C	q _{exp}	Pseudo-second order model				Pseudo-first order model	
		q _e	k ₂ × 10 ⁻³	h ₀ × 10 ⁻⁴	R ²	K ₁ × 10 ⁻³	R ²
15	0.069	0.0855	33.447	0.245	0.9984	334.626	0.9881
25	0.089	0.1052	35.834	0.397	0.9974	391.049	0.9967
35	0.075	0.0836	81.48	0.569	0.9996	514.951	0.9889
45	0.072	0.0828	56.938	0.390	0.9985	441.255	0.9916

q_e: mg/g, h₀: mg/g min, k₁: 1/min, k₂: g/mg min

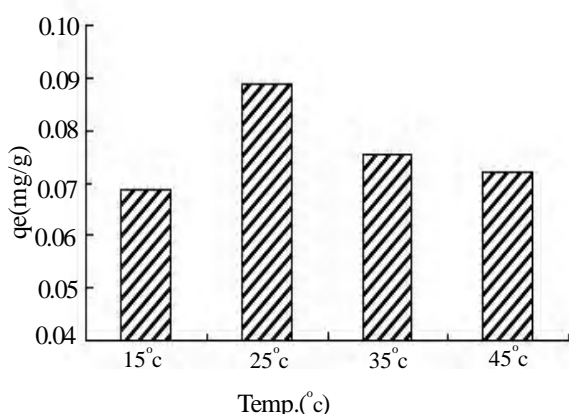


Fig. 6. Effect of temperature on As(III) adsorption by silica ceramic (pH 7.5, adsorbent dose 20 g/L and initial As(III) concentration 2 mg/L)

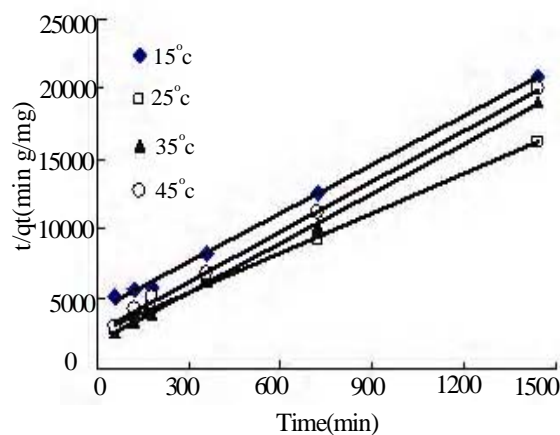


Fig. 7. Pseudo-second order kinetics for adsorption of As(III) onto silica ceramic at different temperature (pH 7.5, adsorbent dose 20 g/L and initial As(III) concentration 2 mg/L)

systems. Several isotherm equations are available and the two popular isotherms are selected in this study, the Langmuir and Freundlich isotherms. The Langmuir adsorption isotherms assumes that adsorption takes place at specific homogeneous sites within the adsorbent and has found successful application to many sorption process of monolayer adsorption. The Langmuir adsorption isotherm can be written as:

$$q_e = \frac{q_m b C_e}{1 + b C_e} \quad (10)$$

The Langmuir parameters were obtained by fitting the experimental data to the linearized equation derived from Eq. (10):

$$\frac{C_e}{q_e} = \frac{1}{b q_m} + \frac{C_e}{q_m} \quad (11)$$

$$\frac{1}{q_e} = \frac{1}{b q_m} \frac{1}{C_e} + \frac{1}{q_m} \quad (12)$$

Where, q_e is the adsorbent amount (mg/g) of the As(III), C_e is the equilibrium concentration of the As(III) in solution (mg/L), q_m is the monolayer adsorption capacity (mg/g) and b is the constant related to the free energy of adsorption (L/mg). Based on Eq. (11) and Eq. (12) the isotherms were fitted to the adsorption data obtained. The Langmuir adsorption exponents for Eq. (11) and Eq. (12), the q_m and b are determined from the linear plots of C_e/q_e versus C_e (Figure not shown) and $1/q_e$ versus $1/C_e$ (Fig. 8) and calculated correlation coefficients for these isotherms, are shown in Table 5. The values of the Langmuir constant were calculated from the slopes and intercepts of the plots. The magnitude of Langmuir constant b is small (0.1077 L/mg) and the adsorption capacity q_m was determined as 1.7886 mg/g. In order to predict the adsorption efficiency of the adsorption process, the dimensionless equilibrium parameter R_L was determined by using the following equation (Magdy and Daifullah, 1988):

$$R_L = \frac{1}{(1 + b C_0)} \quad (13)$$

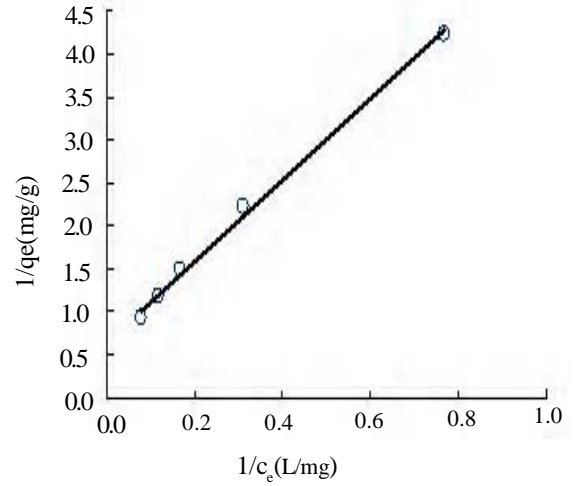


Fig. 8. Langmuir Isotherm for As(III) adsorption onto silica ceramic at different initial concentration

Table 5. Langmuir and Freundlich isotherm constant for adsorption of As(III) by silica ceramic

	Langmuir isotherm		Freundlich isotherm	
Parameters	C_e/q_e	$1/q_e$	Parameters	Value
q_m (mg/g)	1.7886	1.5228	k_f (mg/g)	0.2027
b (L/mg)	0.1077	0.1392	n	1.5147
R^2	0.9853	0.9968	R^2	0.998

Where, C_0 is the initial concentration and b is the Langmuir isotherm constant. The parameter R_L indicates the shape of isotherm accordingly, as explained in Table 6. The (Fig. 9). shows that the R_L values at different initial As(III) concentration indicating a highly favorable adsorption.

Table 6. Separation factor for shape of isotherm

Values of R_L	Type of isotherm
$R_L > 1$	Unfavorable
$R_L = 1$	Linear
$0 < R_L < 1$	Favorable
$R_L = 0$	Irreversible

The Freundlich isotherm is an experimental equation in use to describe heterogeneous systems:

$$q_e = K_f C_e^{\frac{1}{n}} \quad (14)$$

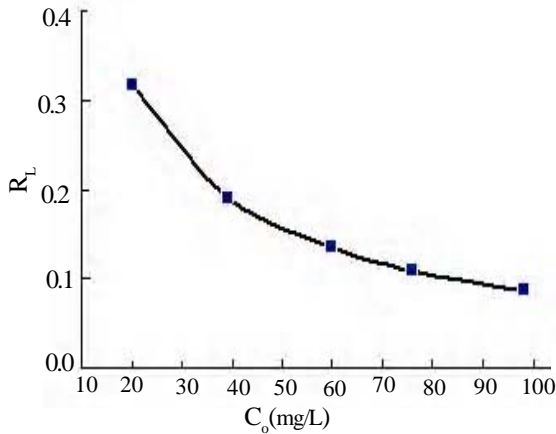


Fig. 9. Separation factor for As(III) adsorption by silica ceramic

The linearized form of Freundlich equation is:

$$\log q_e = \log k_f + \frac{1}{n} \log C_e \quad (15)$$

Where, q_e is the adsorbent amount (mg/g) of the As(III), C_e is the equilibrium concentration of the As(III) in solution (mg/L), K_f is the adsorption capacity of Freundlich constant (mg/g) and n is the adsorption intensity of Freundlich adsorption isotherm constant. The Freundlich adsorption exponents for Eq. (14), the K_f and n are determined from the linear plots of $\log q_e$ versus $\log C_e$, which are shown in Fig. 10 and respective data, are shown in Table 5. It is generally stated that the values of n in the range of 1 to 10 represents good adsorption (Magdy and Daifullah, 1988). In this study the Freundlich constant n (1.5147) was $1 < n < 10$, represents favorable adsorption.

Table 7. Comparison of adsorption capacities of various adsorbents for As(III)

Adsorbent	Adsorption capacity (mg/g)	References
Iron oxide coated sand	0.0286	Gupta <i>et al.</i> , 2005
Activated alumina	0.2249	Singh and Pant, 2004
Activated red mud	0.8838	Altundogan <i>et al.</i> , 2002
Calcined bauxite	1.362	Ayoob <i>et al.</i> , 2007
Silica ceramic	1.7886	This study

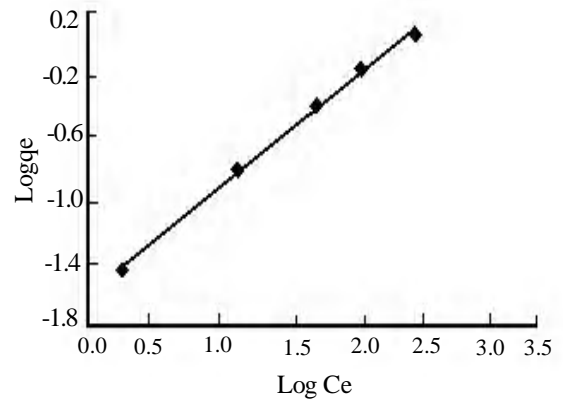


Fig. 10. Freundlich Adsorption Isotherm for As(III) adsorption onto silica ceramic at different initial concentration

As shown in Table 5, Fig. 7 and Fig. 9, the Freundlich equation represents adsorption process very well; the correlation coefficient, R^2 value is higher for the Freundlich isotherm than the Langmuir isotherm, indicating a very good mathematical fit. Earlier some researcher investigated other adsorbents such as activated red mud, iron oxide coated sand, calcined bauxite and activated alumina for the removal of As(III) from aqueous solutions. Table 7 shows the comparison of the maximum monolayer adsorption capacities of various adsorbent of As(III) removal. The value of adsorption capacity, q_m (1.7886 mg/g) in this study is larger than those in most of previous results of different studies. Therefore, the silica ceramic can be use as an effective As(III) removal from contaminated water.

CONCLUSION

Based on the experimental results of this batch study, following conclusions can be drawn:

- Adsorption of As(III) by silica ceramic has been revealed to depend on the initial pH (6 to 7.5), adsorbent amount and initial concentration of solution. The best result of adsorption performed at pH 7.5, but efficiency decreases with the increase of pH.
- Batch experiment showed that silica ceramic could remove a large amount arsenite from aqueous solutions efficiently than mentioned other referenced study.
- The pseudo-second order kinetic model fitted very well with a good correlation and dynamical adsorption behavior of As(III).

- Adsorption equilibrium data follows both Langmuir and Freundlich isotherms model, however Freundlich isotherm model well fitted by the experimental data. The capacity of adsorption 1.7886 mg/g was calculate from Langmuir isotherm at initial pH 7.5 and optimum study conditions. The dimension less separation factor (RL) showed that silica ceramic was favorable for removal of As(III) from aqueous solutions. Silica ceramic can potentially be used an effective and inexpensive adsorbent for arsenite removal from aqueous solutions.

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