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Arsenic(III) Removal from Contaminated Water using Silica Ceramic: A Batch Adsorption Study

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Abstract: The silica ceramic (S-K) has been used as a potential low-cost adsorbent for the removal of As(III) from contaminated water in batch studies. Results showed that an adsorbent dose at 90 g L^{-1} could effectively remove 96% of As(III) from initial concentration of 0.5 mg L^{-1} within 3 h. It was also reveled that As(III) uptake increased with increasing contact time and As(III) concentration decreased with increasing adsorbent dosage. It was observed that As(III) removal is relatively dependent on pH and temperature variations. High adsorption of As(III) was found at pH 7.5 and at 25°C. The adsorption equilibrium data followed both Langmuir and Freundlich isotherms. The Freundlich isotherm provided the best correlation for the adsorption of As(III) onto the silica ceramic. The maximum capacity of adsorption was found (q_m) 1.1041 mg g^{-1} at different concentrations at 40 g L^{-1} fixed adsorption dosage. The results suggest that silica ceramic (S-K) is a potential adsorbent for removal As(III) ions effectively from contaminated water.

Key words: Arsenic, removal, adsorption, batch studies, contaminated water, silica ceramic

INTRODUCTION

Arsenic is a toxic trace element found in natural and industrial waters. In recent years, arsenic contamination of water and groundwater with the effect of toxicity has become an increasing concern on a global perspective. There have been widespread reports of arsenic poisoning in Bangladesh (Chowdhury *et al.*, 2000), India [West Bengal] (Das *et al.*, 1995) and China, Taiwan (Tseng *et al.*, 1968), Vietnam (Berg *et al.*, 2001), United States, Argentina, Chile, Mexico, etc. In natural water, arsenic is primarily present in inorganic forms and exists in two predominant species, arsenate [As(III)] and arsenate [As(V)]. As(V) is the major arsenic species in surface water, while As(III) is the dominant arsenic species in groundwater since it is favored under reducing conditions. As(III) is more toxic to biological systems than As(V) (Ferguson and Devis, 1972; Cullen and Reimer, 1989; Korte and Fernando, 1991).

Inorganic species of arsenic represents a potentials threat to the environment, human health and animal heath due to their carcinogenic and other effects. Long-term uptake of arsenic contaminated water causes liver, lung, kidney, bladder, skin and nerve tissue injuries (Roberts *et al.*, 2004; Thirunavukkarasu *et al.*, 2003). So, it is very important to remove arsenic from wastewater and contaminated drinking water. Usually, a removal technique of arsenic from aqueous system requires: (i) safe operation with respect to the maximum contaminant level, (ii) high efficiency, (iii) easy for application and (iv) low cost (Xu *et al.*, 2002).

Conventional water treatment processes remove toxic metal ions through mechanism such as sorption and particle removal. Advanced water treatment techniques, which can be used as either primary treatment or post treatment, involve ion exchange, reverse osmosis, adsorption, coagulation, precipitation, adsorption-coprecipitation with hydrolyzing metals and so on (McNeill and Edwards, 1997; Hering, 1996; Huang and Vane, 1989; Sorg and Logsdon, 1978). But due to excessive use of chemicals, bulky sludge, high cost, these techniques are not feasible at small-scale or household level. Among these, adsorption currently appears to have the best potential for overall treatment and it can be expected to be useful for a wide range of compounds, more so than any other listed process. Several studies have demonstrated that arsenic removal can be achieved by technologies namely activated alumina sorption, (Singh and Pant, 2004), iron oxide-coated sand (Gupta *et al.*, 2005; Zang and Itoh, 2005; Zeng, 2003;

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Joshi and Chaudhuri, 1996), iron oxide-coated cement (Kundu and Gupta, 2007), coagulation with ferric chloride (Meng *et al*., 2000), pressurized granulated iron particles (Vagliasandi and Benjamin, 1998; Driehaus *et al*., 1998), iron oxide-doped alginate manganese dioxide coated sand (Min and Hering, 1998), polymeric ligand exchange, zeoloite (Xu *et al*., 2002), portland cement (Kundu *et al*., 2004), activated red mud (Genc *et al*., 2003; Aultundogan *et al*., 2002) and zero valent iron.

Porous sorbents are promising in removing arsenic from contaminated water. Present study carried out to evaluate the performance of silica ceramic (S-K) for As(III) removal. The effects of process parameters such as effect of adsorbent dosage, effect of initial concentration, effect of pH, effect of temperature, effect of contact time were investigated. These variables in most sorption systems have significant effects on the sorption dynamics parameters. The Langmuir and Freundlich isotherm models were tested for their applicability. The goal of this research was to investigate the efficacy of using silica ceramic to treat water contaminated by arsenite.

MATERIALS AND METHODS

The study was carried out during November 2006 to February 2007 at the relevant laboratory of Kochi University in Japan.

Adsorbent: The silica ceramic (S-K) was prepared with technical assistant of EKOAIRANDO Co., Ltd. (Kochi, Japan). This ceramic was made from special volcano ash blended with iron named *Akadama* soil and burned at 400°C up to 20 minute in controlled conditions. The raw material was collected from Kanuma City of Tochigi prefecture in Japan. The properties of silica ceramic (S-K) are shown in the Table 1.

Reagents: All reagents were of analytical grade, which were purchased from Nacalai Tesque, Inc. and Kanto Chemical Co., Inc. Japan. A synthetic solution of As(III) was made using standard 1000 mg L^{-1} arsenite [As(III)] solutions and deionized (DW) water without further purification. Standard acid and base solutions (0.1N HCl and 0.1N NaOH) were used for pH adjustments. Further working solutions were freshly prepared from stock solution for each experimental run.

Instrumentals: Arsenic concentration determination were carried out using a highly sensitive and sophisticated instrument, the Analyst 200, Perkin Elmer, model No. -41440, Singapore by using EDL at a wavelength 193.7 nm in order to get a more accurate measurement. The instrument was calibrated by using As(III) standard solution of 6, 3 and 1 μg L^{-1}, which was prepared in diluted HCl. pH measurements were made on a pH meter, Mettler Toledo Model No. -MP120, Switzerland.

Table 1: Physico-chemical prosperities of silica ceramic (S-K)

Properties	Kind/value
Particle form	Sphere
Color	Reddish brown
Size (Spherical mm)	0.42±0.02
Bulk density (g cm^{-3})	0.808
Porosity (%)	69.5
Pore volume (cm^3 g^{-1})	0.752

Batch adsorption studies: 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^{-1}, adsorbent dose of 20 g L^{-1} and incubation temperature at 25°C. The effect of the adsorbent dosages were investigated using 10-100 g L^{-1} of adsorbent at a fixed pH of 7.5 and 25 mL of 0.5 mg L^{-1} As(III) solution in 50 mL flasks. These flasks were shaken for 3h to reach the equilibrium at fixed temperature 25°C. Twenty five milliliter of different known initial As(III) concentrations 20, 39, 60, 76, 98 mg L^{-1} were used to investigate the effect of contact time and initial As(III) concentration on the As removal by 80 g L^{-1} of adsorbent at pH 7.5 and 25°C. Samples of the ceramic adsorbent were placed in labeled round-bottomed 50 mL flasks. The sample were collected by predetermine time interval. Effects of temperature (15, 25, 35 and 45°C) were studied with an initial arsenic concentration of 2 mg L^{-1} and adsorbent dosage of 20 g L^{-1}; pH was kept at 7.5 and solution of 50 mL. Adsorption isotherm studies were conducted with 25 mL of varying initial As(III) concentrations (2, 10, 18, 28, 37 and 44 mg L^{-1}), fixed adsorbent dose of 40 g L^{-1} at pH 7.5 and 25°C.

In each experiment, the flasks were sealed with airtight cap and samples were shaken on the mechanical shaker at 150 rpm. Parameters, e.g., adsorbent dosage, contact time, pH and various initial concentration, temperature were optimized for maximum adsorption by the method of continuous variation following batch adsorption experiments mentioned above.

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:

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

Where q_e (mg g^{-1}) is the amount of As(III) anion adsorbed per gram of the ceramic, C_0 (mg L^{-1}) the initial concentration of the solution of As(III), C_e (mg L^{-1}) the equilibrium concentration of the solution of As(III), V (L) the volume of the solution and M(g) the mass of the adsorbent.

The percentage of As(III) removed by the silica ceramic shows the efficacy of removal. It is determined from the ratio of concentration of As(III) present in the solution and particulate phases using the equation:

$$R_e = \frac{C_0 - C_e}{C_0} \times 100 \qquad (2)$$

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 AND DISCUSSION

Effect of pH: In order to determine the most favorable pH for adsorption of As(III), the study was carried out with different pH values as presented in Fig. 1. In this experiment, the duration of arsenic adsorption was kept 60 h. Adsorption isotherms have been prepared from pH 4.0 to 10.7. From Fig. 1 it is clear that maximum adsorption was achieved in the pH of 7.5 for initial As(III) concentration of 2 mg L^{-1}. That means favorable adsorption took place at pH 7.5, same as Altundogan *et al.* (2002) and therefore all further experiments were carried out at pH of 7.5. Low adsorption was observed at higher and lower pH values.

The pH of the solution determines the concentrations distribution of the ionic forms of As(III). The removal of As(III) by iron-coated sand (Gupta *et al.*, 2005; Thirunavukkarasu *et al.*, 2003), amorphous ferric hydroxide (Wilkie and Hering, 1996) nearly similar trends were reported.

Effect of contact time and initial As(III) concentration: To evaluate the effect of initial As(III) concentration (C_0) on adsorption, studies were conducted with initial As(III) concentrations (C_0) of 98, 76, 60, 39 and 20 mg L^{-1} with silica ceramic (S-K) as the adsorbent material.

Based on the data, plots were prepared between the adsorbed amount of As(III) vs time. Figure 2 described these plots at five different C_0 values. These plots indicate that As(III) adsorbed increase rapidly in the beginning and very slowly towards the end of run. In other words, it can be observed from Fig. 2 that the concentration of As(III) adsorbed increased with time and at some points of time reached a constant value beyond which no more As(III) was removed from the solution.

From Fig. 2, the equilibrium time was found to be 12 h for 20 mg L^{-1}, 24 h for 39 mg L^{-1} and 36 h for 60, 76 and 98 mg L^{-1}. The amount of As(III) adsorbed at equilibrium (q_e) increased from 0.22 to 1.0 mg g^{-1} as the concentration was increased from 20 to 98 mg L^{-1}. It can be concluded that high removal at low concentration is important in terms of industrial application. The adsorption curves are single, smooth and continuous leading to saturation, suggesting the possible monolayer coverage of As(III) ions on the surface of adsorbent.

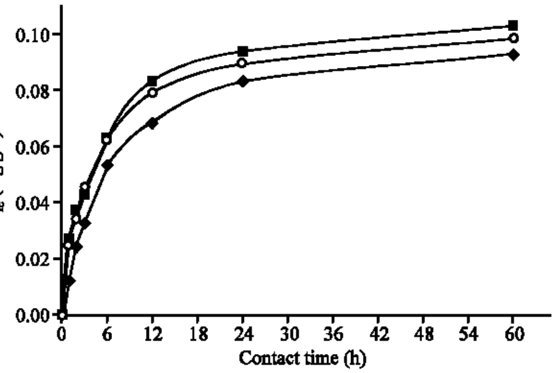

Fig. 1: Effect of pH on As(III) removal on silica ceramic S-K. Conditions: contact time, 60 h; adsorbent dosages 20 g L^{-1}; As(III) concentration, 2 mg L^{-1}; temperature, 25°C; solution pH, (—◆—) pH 4.0, (—■—) pH 7.5 and (—○—) pH 10.7

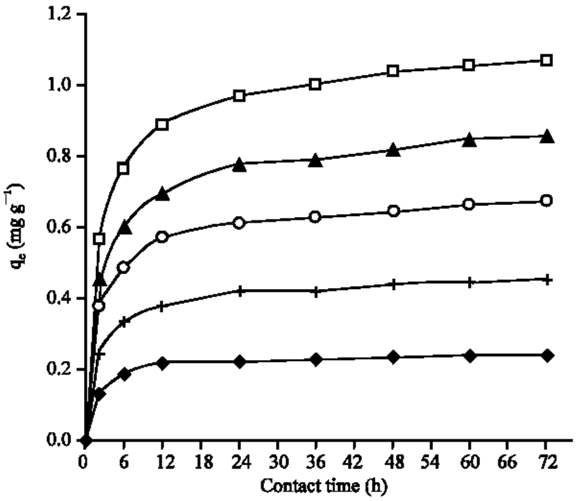

Fig. 2: Arsenic removal by silica ceramic (S-K) as a function of different As(III) concentrations. Initial condition: pH 7.5, dosage 80 g L^{-1} and temperature 25°C. (□) 98 mg L^{-1}, (▲) 76 mg L^{-1}, (○) 60 mg L^{-1}, (+) 39 mg L^{-1} and (◆) 20 mg L^{-1}

Effect of adsorbent dosage: The effect of adsorbent dosage on As(III) uptake is described in Fig. 3, which shows that adsorption efficiency increased very rapidly with an increase in adsorbent dosage 10 to 50 g L^{-1}. At 50 g L^{-1} As(III), 82.3% removal efficiencies was observed. The higher uptake of As(III) with increase in the adsorbent dosage up to 90 g L^{-1} was observed and thereafter remained nearly constant.

However, it was found that the adsorption capacity q_e decreases with the increase of adsorbent dosage. 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).

Effect of temperature: The temperature used for experiment was varied from 15 to 45°C. Figure 4 shows the effect of temperature on removal of As(III) there was no significant change in removal efficiency at different temperature range. The adsorption of arsenic on silica ceramic (S-K) was highest at 25°C.

The results indicate that a rise in the temperature caused the increase of removal from 0.075 to 0.095 mg g^{-1} up to 25°C and then decreased up to 35°C and thereafter remained almost constant. This result revealed that the process is slightly endothermic. Therefore other experiments were conducted at 25°C.

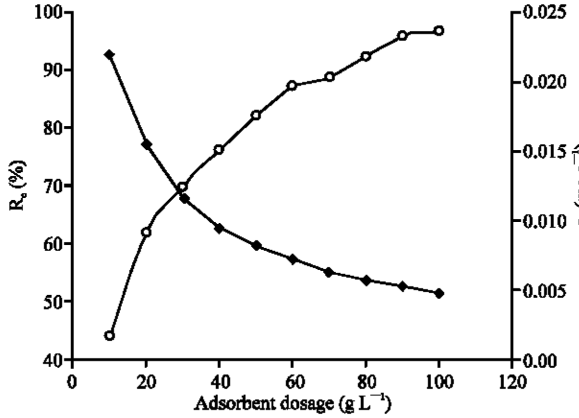

Fig. 3: Effect of adsorbent dosage on As(III) adsorption. Conditions: Initial As(III) concentration, 0.5 mg L^{-1}; pH 7.5, temperature 25°C, contact time 3 h. (○) % of As(III) removal, R_e and (◆) adsorbed amount, q_e(mg g^{-1})

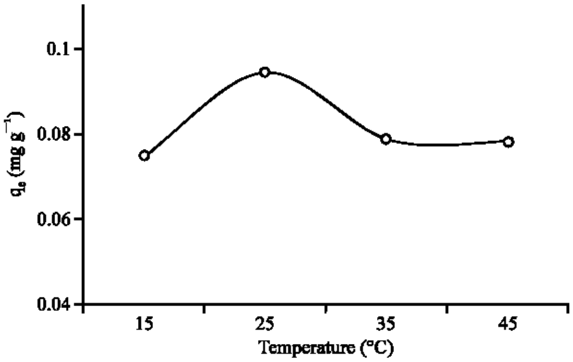

Fig. 4: Effect of temperature on As(III) adsorption. Initial conditions: Initial pH 7.5, adsorption dosage, 20 g L^{-1}; Initial As(III) concentration, 2 mg L^{-1}; temperature, 15, 25, 35 and 45°C; equilibrium time 48 h

Adsorption isotherm: The plots of arsenic uptake against equilibrium concentration (Fig. 5) indicate that adsorption increased initially with concentration but then reached the saturation point. Langmuir and Freundlich isotherms were applied to study the adsorption equilibrium. The Langmuir isotherm is based on assumption that (i) maximum adsorption corresponds to a saturated monolayer of adsorbate molecules on the adsorbent surface, (ii) that the energy of adsorption is constant and (iii) that there is no transmigration of adsorbate molecules in the plane of adsorbent surface.

The Langmuir isotherm is expressed as:

$$q_e = q_m \frac{bC_e}{(1+bC_e)} \tag{3}$$

and its linearized form is

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \frac{C_e}{q_m} \tag{4}$$

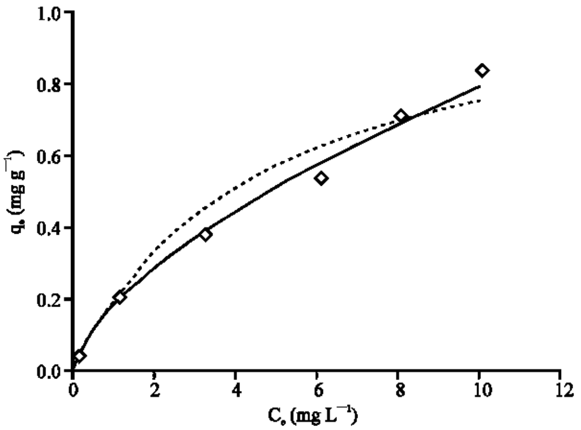

Fig. 5: Arsenic adsorption on silica ceramic (S-K) at 25°C with following conditions: pH 7.5, Contact time 60 h, dosage 40 g L^{-1}, initial As(III) concentration is 2 to 44 mg L^{-1}, (◇) data point, (- - -) Langmuir isotherm curve, (——) Freundlich Isotherm curve

Where q_e is adsorbed amount (mg g^{-1}), C_e is equilibrium arsenic concentration on in solution (mg L^{-1}), q_m is monolayer capacity of the adsorbent (mg g^{-1}) and b is adsorption constant or energy of adsorption (L mg^{-1}).

The Freundlich isotherm equation is expressed as:

$$q_e = k_f C_e^{\frac{1}{n}} \quad (5)$$

and its linearized form is

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

Where q_e is adsorbed amount (mg g^{-1}), C_e is equilibrium arsenic concentration on in solution (mg L^{-1}), K_f (mg g^{-1}) is the Freundlich constant related to adsorption capacity and n (dimensionless) is constant related to energy of intensity of adsorption.

Adsorption isotherms were obtained in terms of Eq. 4 and 6 by using experimental adsorption results in these equations. The Langmuir and Freundlich exponents q_m, b, K_f and n are determined from the linear plots of C_e/q_e vs C_e and log q_e vs log C_e which are shown in Fig. 6 and 7, respectively and respective data are shown in Table 2.

As shown Table 2, the Freundlich equation represents the 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. The fact that the Freundlich isotherm fits the experimental data very well (Fig. 5) may be due to homogeneous distribution of active sites on the ceramic surface. The maximum adsorption capacity obtained from Langmuir model was found 1.1041 mg g^{-1}. Previously some studies investigated other adsorbents such as activated red mud, iron oxide coated sand, calcined bauxite and activated alumina for the removal As(III) aqueous solution. A comparison of the results obtained in this study with those in the previously reported works are given Table 3, which shows the adsorption capacities of various low-cost adsorbents in aqueous solution for As(III) removal. It can be stated that the findings of this study is quite good compare to the other studies.

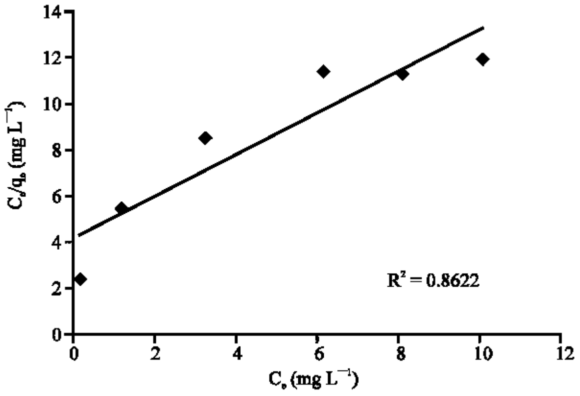

Fig. 6: Langmuir plots for As(III) adsorption by silica ceramic (S-K). Initial conditions: pH 7.5, Contact time 60 h, dosage 40 g L^{-1}, initial As(III) concentration 2 to 44 mg L^{-1}

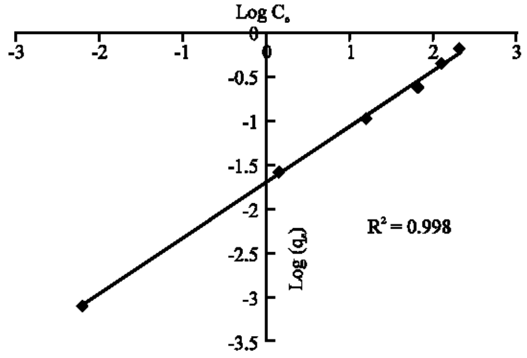

Fig. 7: Freundlich plots for As(III) adsorption by silica ceramic (S-K). Initial conditions: pH 7.5, Contact time 60 h, dosage 40 g L^{-1}, initial As(III) concentration 2 to 44 mg L^{-1}

Table 2: Langmuir and Freundlich isotherm constant for adsorption of As(III) by silica ceramic (S-K)

Langmuir		Freundlich	
Constant	Value	Constant	Value
q_m	1.1041	k_f	0.1836
b	0.2151	n	1.5710
R^2	0.8622	R^2	0.9980

q_m (mg g^{-1}): Maximum amount of As(III) adsorbed per gram of silica ceramic; b (L mg^{-1}): Langmuir adsorption constant; K_f (mg g^{-1}): Freundlich adsorption constant measures adsorption capacity; n: Freundlich constant measures strength of adsorption; R^2: Correlation coefficient

Table 3: Comparison of As(III) adsorption of silica ceramic(S-K) with other adsorbents

Adsorbent	Adsorption capacity (mg g^{-1})	Dose (g L^{-1})	Initial As(III) concentration (mg L^{-1})	References
Activated red mud	0.8838	20	25-30	Altundogan *et al.* (2002)
Calcined bauxite	1.3620	-	0.5-8	Ayoob *et al.* (2007)
Iron oxide coated sand	0.0286	20	0.1-0.8	Gupta *et al.* (2005)
Activated alumina	0.2249	1-13	1	Singh and Pant (2004)
Silica ceramic (S-K)	1.1041	40	2-44	Present study

CONCLUSIONS

This study revealed that silica ceramic can be used as an efficient adsorbent for As(III) removal in contaminated water treatment. The adsorption was found to be dependent on pH, adsorbent dosage and contact time. Best results were obtained at pH values 7.5 and the efficiency decreases with the lower and higher pH. As(III) was rapidly adsorbed when lower concentrations were used. The amount of As(III) uptake (mg g^{-1}) was found to increase with the increase in contact time and initial As(III) concentration, but decreased with an increase in adsorbent dosage. The maximum adsorption was found at 25°C temperature.

The adsorption isotherm follows Freundlich isotherm model better than Langmuir isotherm. The findings of the study revealed that silica ceramic has high potential for use the in the removal of arsenate from contaminated water, but further work is needed to increase the arsenate adsorption capacity of silica ceramic.

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