



Arsenic removal by iron oxide coated sponge: Experimental performance and mathematical models

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ABSTRACT

Millions of people worldwide are at risk from the presence of arsenic in groundwater. In this study, adsorption equilibrium and long term experiments were carried out to evaluate the performance of iron oxide coated sponge (IOCSp) in arsenic removal. It was found that maximum adsorption capacity of IOCSp for As(III) and As(V) calculated by Sips isotherm was 4.2 and 4.6 mg/g of IOCSp, respectively. A filter packed with small amount of 25 g IOCSp maintained a consistent arsenic removal efficiency of 95% from synthetic solution containing arsenic concentration of as high as 1000 µg/L. This produced a throughput volume of 153 and 178 L of water containing As(III) and As(V), respectively before any need for regeneration or disposal of IOCSp. IOCSp could be regenerated by washing it with NaOH solution. The dynamic (column) adsorption kinetics were successfully predicted by the Thomas and Nikolaidis modified models.

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1. Introduction

Arsenic (As) is a toxic semi-metallic element that can be fatal to human health. Arsenite [As(III)] and arsenate [As(V)] are the two predominant arsenic species found in groundwater [1]. Arsenic can have a significant adverse impact on the environment. Arsenic pollution in water has been found in many parts of the world, especially in developing countries such as Bangladesh, India, Nepal, and Vietnam [2,3]. The arsenic contamination in water forced the water and health authorities to introduce stringent standards for arsenic levels in drinking water. World Health Organization (WHO) has recommended the arsenic level in drinking water to be 10 µg/L. Rigorous criteria of arsenic level have been enforcing water authorities to identify and put into practice suitable and cost-effective arsenic removal technologies.

Since adsorption using a fixed bed treatment system is simple for operation and handling whilst having a good regeneration capacity and sludge free operation, this technology has become very popular for arsenic removal in small-scale treatment systems. A number of adsorbents such as granular ferric hydroxide, activated alumina, and red mud have been used to remove arsenic from water [4–9].

Thirunavukkarasu et al. [4] used granular ferric hydroxide (GFH) (which consists of ferric oxihydroxide of approximately 52 to 57%

by mass and moisture content of 43–48%) for arsenic removal. They found that the adsorption capacity for As(III) and As(V) was 0.11 mg/g and 0.16 mg/g respectively. Some researchers used activated alumina for arsenic adsorption [5–7]. The results from Singh and Pant [5] showed that adsorption capacities of As(III) on activated alumina depended on temperature and this value reduced from 0.18 mg/g at 25 °C to 0.145 mg/g at 35 °C. As(III) and As(V) adsorption on red mud was reported by Altudogan et al. [8,9]. They found that As(III) was effective removed in the alkaline environment (pH 9.5) whereas As(V) removal was effective in the acidic range (1.1–3.2). The arsenic removal capacities were 0.66 mg/g for As(III) and 0.51 mg/g for As(V). The arsenic adsorptive capacity of red mud increased when it was treated by heat and acid [9]. Fuhrman et al. [10] used chemically modified red mud (Bauxsol) or activated Bauxsol (AB) coated sand, to remove As(V) from water. Bauxsol-coated sand (BCS) and AB-coated sand (ABCS) were prepared by mixing Bauxsol or AB with wet sand and drying. They reported that adsorption capacity for BCS was 3.32 and 1.64 mg/g at pH 4.5 and 7.1, respectively. Arsenic adsorption on ABCS was 2.14 mg/g at pH 7.1.

In adsorption, the surface area exposed plays an important role in adsorption efficiency of adsorbents. The adsorption rate of nanoscale zerovalent iron (1–120 nm diameter) for As(III) was about 1000 times faster than that of micron-sized iron [11]. The maximum adsorption of As(III) on nZVI was calculated by Freundlich adsorption isotherm as 3.5 mg/g.

Some researchers tried to coat iron oxide on different materials and used them to remove arsenic. Gupta et al. [12,13] tried to use both iron oxide coated and uncoated sands. They used 20 g/L of

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fixed adsorbent dose for removing As(III) from synthetic solution containing initial As(III) concentrations of 100–800 $\mu\text{g/L}$. The maximum adsorption capacity of As(III) onto iron oxide coated sand was five times higher (0.029 mg/g) than that onto uncoated sand (0.006 mg/g) at pH 7.5 in 2 h. Kundu et al. [14–16] utilized an iron oxide treated cement (IOCC) as an arsenic adsorbent. They found that the uptake of As(III) is very rapid and most of fixation occurred within the first 20 min [14]. The As(III) adsorption capacities calculated by Langmuir isotherm were 0.73, 0.69, 0.67 and 0.66 mg/g at 15 °C, 25 °C, 35 °C and 45 °C, respectively [15]. The adsorption capacity of IOCC with As(V) was found to be much higher than that of As(III). The values were 3.39 mg/g at 15 °C, 3.86 mg/g at 25 °C and 4.63 mg/g at 35 °C [16].

Sponge is seen to be a promising material for adsorption because it is non-toxic, has high internal porosity and specific surface that facilitate arsenic absorption. In addition, sponge can be easily compacted to a very small volume to facilitate disposal. Importantly, sponge is cost-effective so it can be applied in small rural communities as well as in developing countries. To achieve a high arsenic adsorption, sponge should be coated with iron oxide. After adsorption of arsenic onto a sponge matrix, the sponge can be safely disposed of. Therefore, a material namely iron oxide coated sponge (IOCSp) has been developed by coating iron oxide on polyurethane sponge [17,18].

Our earlier pilot study found that a 180 g of IOCSp packed in a bucket (200 mm in diameter and 350 mm in height) could reduce arsenic concentration of 480 $\mu\text{g/L}$ in 1500 L contaminated natural water to below 40 $\mu\text{g/L}$ [17]. Another short term IOCSp column experiments to evaluate the performance of the IOCSp column was conducted with contaminated groundwater (56–156 $\mu\text{g/L}$) in Kelliher (Canada) and in Hanoi (Vietnam). Even a small amount of IOCSp of 8 g could treat up to 63 L (bed volume of 310) of Kelliher groundwater (Canada) and 75 L (bed volume of 390) of Hanoi groundwater (Vietnam) to a value less than 18 and 50 $\mu\text{g/L}$, respectively.

Our previous results on batch kinetics of IOCSp with arsenic showed that the pH in the range of 6–8 did not have any significant variation on the removal of As(III) and As(V). After 9 h, 0.15 g of IOCSp could removed 93–96% of both As(III) and As(V) in synthetic solution containing 260 $\mu\text{g/L}$ of arsenic in 3 experimental pH of 6, 7, and 8 [18].

The effect of temperature on the adsorption equilibrium and batch kinetics of IOCSp with As(III) were investigated at four different temperatures (15 °C, 25 °C, 35 °C and 45 °C) [19]. Under the steady-state reaction conditions, the change in Gibbs free energy (ΔG°) from –14.42 to –15.92 kJ/mol, and enthalpy (ΔH°) of –18.14 kJ/mol together with entropy change (ΔS°) of 0.05 kJ/mol/K proved higher temperature was unfavourable spontaneous reaction for As(III) adsorption by IOCSp. This result was similar to the adsorption of As(III) on iron oxide coated cement where the adsorption capacity decreased when the temperature increased [15]. The adsorption capacity of IOCSp with As(III) calculated by Langmuir were 4.16, 3.85, 3.81, and 3.63 mg/g at 15 °C, 25 °C, 35 °C and 45 °C respectively. Thermodynamics calculations also indicated that the adsorption of As(III) on the adsorbents was exothermic reaction [19].

The modeling of the arsenic adsorption in the column packed with IOCSp will help to estimate mass transfer coefficient and investigate the design factors. A number of models have been used in adsorption [20–24]. The Thomas model can easily be applied to determine the adsorptive capacity of the adsorbent and to study the adsorption kinetics. Further, this model provides an approach for the design and projection of the performance of fixed bed absorbers without the need to conduct extensive experimental work and mathematical formulation.

This research evaluated the performance of iron oxide coated sponge in removing both As(III) and As(V) in drinking water. Regen-

Table 1
Water quality parameters.

	Parameters	Concentration
1.	Arsenic	1–5
2.	Iron	0.07
3.	Manganese	0.01
4.	Nitrate	0.74
5.	Turbidity (NTU)	0.2
6.	Copper	0.005
7.	Zinc	<0.005
8.	Phosphate	0.004
9.	Silicate	0.8

Note: All parameters except pH and turbidity are in mg/L.

eration of the exhausted IOCSp was also investigated. The modeling of transport of arsenic in the columns packed with IOCSp was also assessed by 2 different models namely Thomas model and Niko-laidis modified model.

2. Materials and methods

2.1. Water

The experiments were conducted with synthetic solution. The synthetic solution used for dynamic experiments represents high arsenic contamination in water of 1000 $\mu\text{g/L}$. The composition of the synthetic solution used in this study is presented in Table 1.

2.2. Iron oxide coated sponge (IOCSp)

The sponge which was used in this study is a commercial polyurethane sponge. The dry density of polyurethane sponge is 28 kg/m³, average pore area is 0.7 mm². The IOCSp contains 12% of iron oxide. The method of preparation of iron oxide coated sponge is explained elsewhere [18].

An experimental study was made to study the specific surface area (S_{BET}) and porosity of IOCSp. The results were difficult to interpret due to the sample size. However, a previous study of Moe and Irvine [25] showed that the surface area of polyurethane sponge was 620 m²/m³ and porosity at 65% moisture content was 0.85.

2.3. Batch equilibrium study

Equilibrium studies were conducted at the room temperature of 22 °C. In these experiments, different amounts of adsorbent (0.018 g to 1.296 g) was placed into 250 mL Erlenmeyer flasks containing 100 mL of the 5000 $\mu\text{g/L}$ of As(III) or As(V). The flasks were shaken at 130 rpm for 20 h. After 20 h of contact time, samples from each flask were decanted and analyzed for the residual arsenic in the solution.

2.4. Column study

Experiments were conducted to study the removal of both As(III) and As(V) in synthetic solution. Column experiments were conducted at the normal pH (between 6.5 and 7.3). In the experiment, a glass column of 45 mm diameter and 940 mm height was used in this study. The water was run through the packed column in the upflow direction using a peristaltic pump at a filtration velocity of 0.17 m/h. The column was packed with 25 g (1240 mL, fixed bed height = 78 cm) of IOCSp. Samples from the column tests were collected at regular time intervals and analysed for residual arsenic by using a hydride generation atomic absorption spectrometry (HGAAS), model AAS 932 Plus and HGGBC 3000. After 43 days running, the IOCSp was regenerated with 12 L of 0.3 M NaOH and backwashed with deionized water until the pH of the effluent

was equal to the pH of the influent. Then the columns were run again.

2.5. Modeling

Adsorption equilibrium of IOCSp with arsenic can be described by Langmuir, Freundlich and Sips isotherms. The Langmuir isotherm is given by:

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

where C_e is the equilibrium concentration (mg/L), b is a Langmuir constant related to the binding energy of adsorption (L/mg), and q_m is the saturated maximum monolayer adsorption capacity (mg/g).

The Freundlich isotherm is an empirical equation developed based on the assumption that the adsorbent has a heterogeneous surface composed of different classes of adsorption sites, and each site can be modeled by the following equation:

$$q_e = K_F C_e^{1/n} \quad (2)$$

where K_F is a Freundlich constant indicative of the adsorption capacity of the adsorbent, and n is an experimental constant indicative of the adsorption intensity of the adsorbent.

The Sips model is another empirical model representing equilibrium adsorption data. This isotherm model has the features of both the Langmuir and Freundlich isotherm models. As a combination of the Langmuir and Freundlich isotherm models, the Sips model contains three parameters, q_m , b and n , which can be evaluated by fitting the experimental data. For single solute equilibrium data, the Sips adsorption isotherm model can be written as follows:

$$q_e = \frac{q_m (b C_e)^{1/n}}{1 + (b C_e)^{1/n}} \quad (3)$$

The Thomas model as shown in Reynolds and Richards [20] is as follows:

$$\frac{C_{ef}}{C_{if}} = \frac{1}{1 + \exp[k_T / Q_v (q_m m - C_o V)]} \quad (4)$$

where C_{ef} is the effluent adsorbate concentration (mg/L), C_{if} is the influent adsorbate concentration (mg/L), k_T is the Thomas rate constant (L/min mg), q_m is the maximum solid phase concentration of the solute (adsorption capacity) (mg/g), m : mass of the adsorbent (g), V is the throughput volume (L), Q_v is the volumetric flow rate (L/min).

Nikolaidis et al. [21] used another model to describe the transport of arsenic in the columns packed with zero valent iron. In this model, surface adsorption was assumed to be a fast reaction that reaches equilibrium in a fraction of the hydraulic detention time of the column. Arsenic precipitation and co-precipitation in solution or diffusion through the iron oxides (corrosion products) and surface precipitation are much slower processes and can be modeled as a first-order-loss reaction.

$$\frac{\partial C}{\partial t} = -u_x \frac{\partial C}{\partial x} + D_x \frac{\partial^2 C}{\partial x^2} - \frac{\partial S}{\partial t} \frac{\rho_s (1 - n_N)}{n_N} - k_N C \quad (5)$$

where C is the solute arsenic concentration (mg/L), S is the amount sorbed onto porous medium = $K_d C$ (mg/mg), K_d is the linear equilibrium partitioning coefficient (L/mg), ρ_s is the solid density of the particles (mg/L), n_N is the effective porosity, (dimensionless), k_N is the first-order mass transfer loss coefficient (1/s), D_x is the dispersion coefficient in x direction (m^2/s) = αu_x , u_x : the velocity of water (m/s), α : the dispersivity (m).

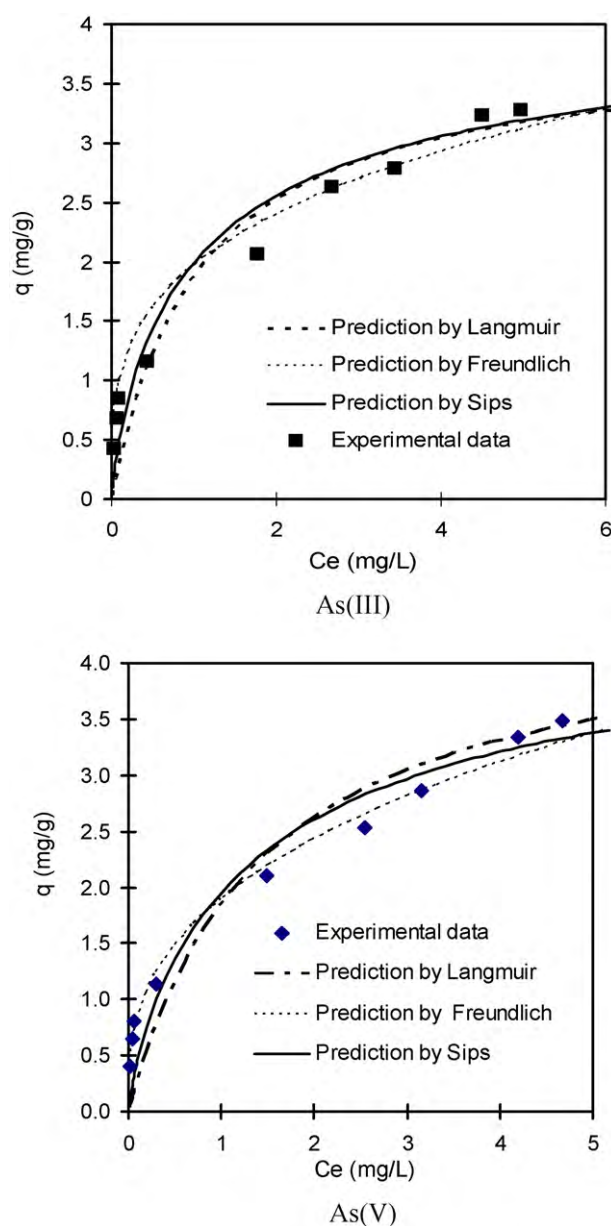


Fig. 1. Prediction of equilibrium adsorption of As by different adsorption models (contact time = 20 h, mixing rate = 130 rpm, temperature = 22 °C).

3. Results and discussion

3.1. Equilibrium adsorption experiments

Adsorption equilibrium experiments were conducted to evaluate the adsorption capacity of IOCSp. The equilibrium results were then fitted with Langmuir, Freundlich, and Sips isotherm equations. The adsorption curves simulated by all of these models fitted well with the observed values. The simulated and the experimental concentrations of the remaining As(III) and As(V) are shown in Fig. 1. The model equations and the isotherm parameters are shown in Table 2.

It can be seen that the Langmuir and Sips models yielded nearly the same values for arsenic adsorption capacity (q_m). The values were 3.85–4.2 mg As(III)/g IOCSp and 4.5–4.6 mg As(V)/g IOCSp respectively. These values were higher than that of a number of adsorbents such as granular ferric hydroxide (0.11 mg/g for As(III) and 0.16 mg/g for As(V)) [4], iron oxide coated sand (0.029 mg

Table 2
Isotherm equations for arsenic removal using IOCSp for synthetic solution.

Parameters	Arsenic species	
	As(III)	As(V)
Langmuir		
q_m	3.85	4.5
b	0.95	0.7
r	0.966	0.974
Freundlich		
k_F	1.97	1.9
n	3.5	2.8
r	0.985	0.990
Sips		
q_m	4.2	4.6
b	0.91	0.74
n	1.27	1.22
r	0.965	0.972

As(III)/g [13], activated alumina (0.18 mg As(III)/g) [5], red mud (0.66 mg/g for As(III) and 0.51 mg/g for As(V)) [8], nanozero valent iron (3.5 mg As(III)/g) [11] and iron oxide coated cement (0.69 mg/g for As(III) and 3.86 mg/g for As(V)) [15,16].

The values of b for both models were almost same. It was in the range of 0.91–0.95 for As(III) and 0.7–0.74 for As(V). This prediction shows that there is not much difference between these two models in describing the adsorption of a single component system. The low value of $1/n$ (<0.36) in the Freundlich isotherm suggests that any large change in the equilibrium concentration of soluble arsenic would not result in a significant change in the amount of arsenic sorbed by the IOCSp. The correlation coefficient (r) for all the isotherms ranged from 0.965 to 0.99, representing a good fit of the observed data.

3.2. Long term performance of IOCSp adsorption column with synthetic solution

The results on the removal of As(III) and As(V) from synthetic solution are presented in Fig. 2. The results showed that the columns could be operated for a long time (24 days and 28 days with As(III) and As(V), respectively) with an effluent arsenic concentration of less than 50 $\mu\text{g/L}$ even with a small amount of IOCSp of 25 g. The throughput volume of As(V) and As(III) was approximately 178 L and 153 L, respectively (which correspond to bed volumes of 144 with As(V) and 123 with As(III)).

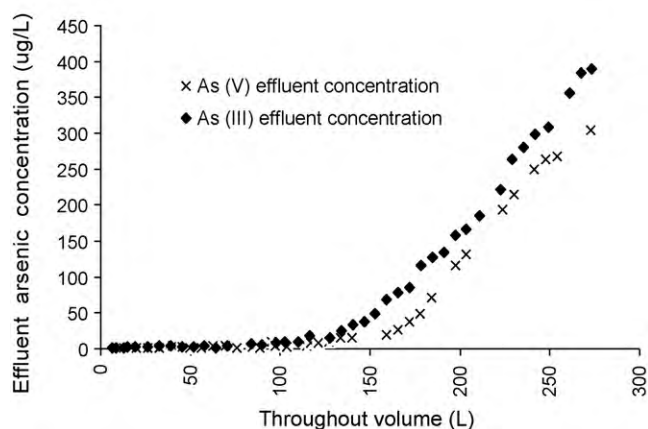


Fig. 2. Removal of arsenic from synthetic solution by IOCSp column (initial As concentration = 1000 $\mu\text{g/L}$; weight of IOCSp = 25 g).

The results of the column studies showed that the performance of IOCSp in terms of bed volume was about 64–70% of iron oxide coated sand (IOCS) reported earlier by Joshi and Chaudhuri [26]. However, the density of IOCS is 74 times higher than that of IOCSp. Thus, in terms of the weight of adsorbent, the performance of IOCSp was about 47–52 times better than IOCS.

Analysis of effluent samples collected during the column test experiment showed that the iron concentrations ranged between 0.03 and 0.1 mg/L, and were nearly the same as the concentrations in the influent.

3.3. Effect of regeneration on arsenic removal efficiency

After 43 days running, IOCSp in the column was regenerated using 0.3 M NaOH at a flow rate of 0.15 m/h until the arsenic concentrations in the effluent were below 50 $\mu\text{g/L}$. The column was then washed again with DI water until the pH of the effluent was equal to the pH of the influent. After washing with DI water, the column was run again with the same flow rate of 0.17 m/h. The results on the removal of arsenic from the regenerated IOCSp column are presented in Fig. 3.

Fig. 3 The results showed that after the regeneration of IOCSp by NaOH solution, the columns could remove both As(III) and As(V) to the value of less than 50 $\mu\text{g/L}$ with the throughput volume approximately 124 L and 147 L respectively (which correspond to bed volumes of 100 with As(III) and 118 with As(V)). These values were about 82% in comparison with the original IOCSp. This showed that IOCSp could be regenerated and used again.

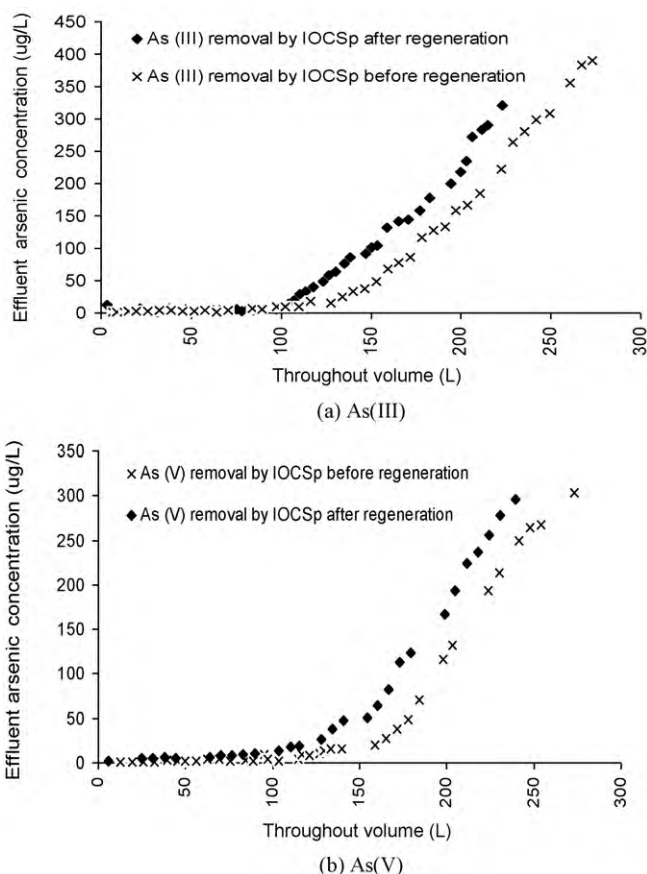


Fig. 3. Removal of arsenic from synthetic solution by regenerated IOCSp column (initial As concentration = 1000 $\mu\text{g/L}$; weight of IOCSp = 25 g).

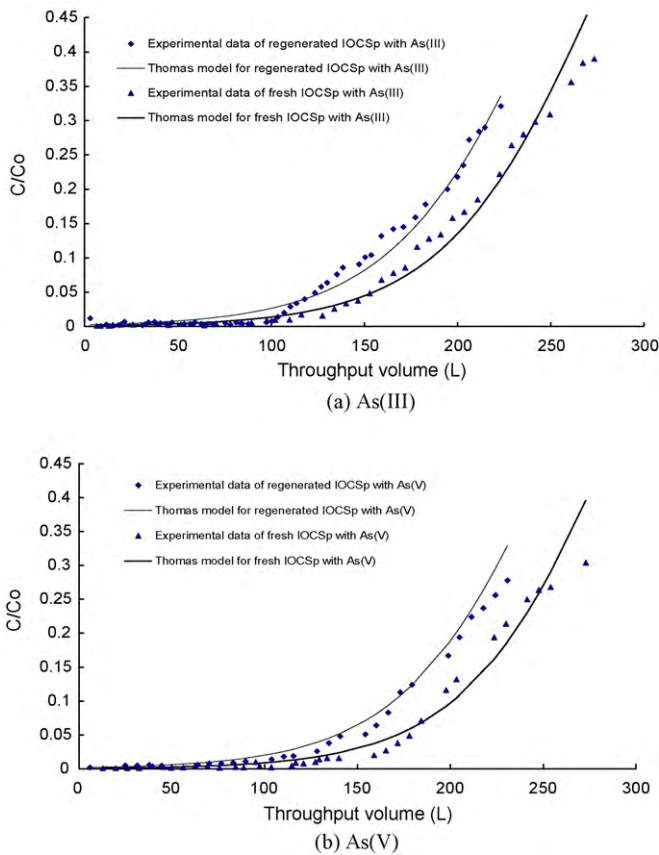


Fig. 4. Experimental and simulated arsenic profiles by the Thomas model in the IOCSp column studies (initial As concentration = 1000 µg/L; IOCSp weight = 25 g).

3.4. Mathematical modeling of IOCSp filter

Since adsorption is the main phenomenon of IOCSp to remove arsenic, Thomas and Nikolaidis adsorption models can be used.

3.4.1. Thomas model

Fig. 4 presents the experimental bed volume dependent breakthrough curves and model simulations. The data fitted quite well with the simulated data of Thomas model. The parameters of the model are presented in Table 3.

The maximum solid phase concentration (q_m) for regenerated IOCSp was about 90% in comparison with fresh IOCSp (10.08 mg As(III) and 10.4 mg As(V) for a gram of regenerated IOCSp). It is thus clear that regenerated IOCSp did not have any significant decrease in removal as compared to the fresh IOCSp.

The results showed that there was not much difference between the breakthrough times calculated from the Thomas model and from experimental data for fresh IOCSp and regenerated IOCSp (Table 4). Here, the breakthrough time is corresponding to effluent concentration 50 µg/L.

As is evident from Table 4, the theoretical and experimental values are comparable with each other.

Table 3
Parameters of Thomas model.

No	Parameters	Unit	Fresh IOCSp		Regenerated IOCSp	
			As(III)	As(V)	As(III)	As(V)
1	q_m	mg/g	11.08	11.6	10.08	10.4
2	k_T	L/min mg	1.06×10^{-7}	1.09×10^{-7}	1.04×10^{-7}	1.07×10^{-7}
3	r		0.976	0.959	0.983	0.993

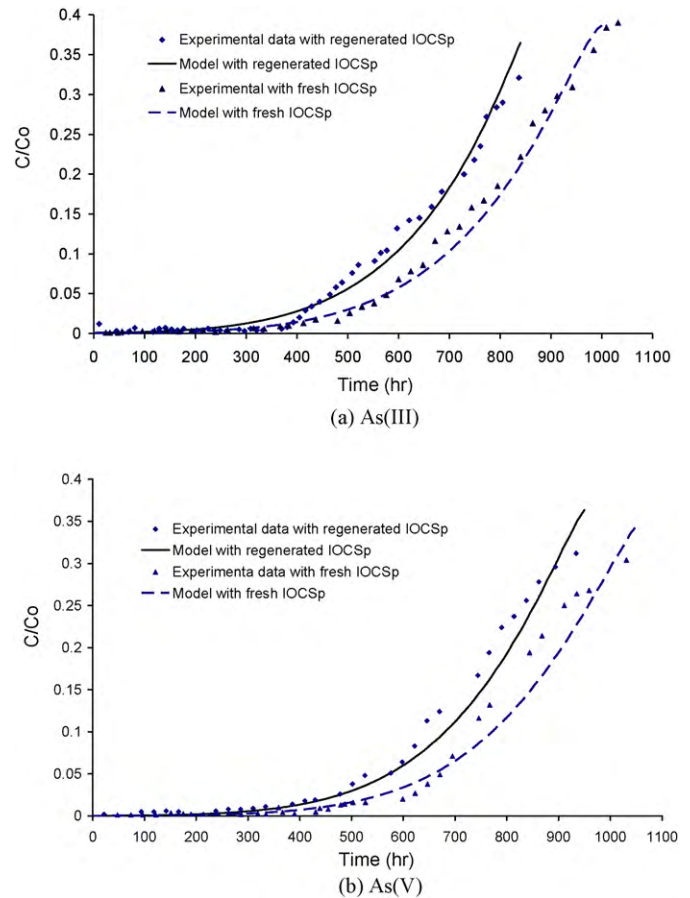


Fig. 5. Experimental and simulated arsenic values by the Nikolaidis modified model in the IOCSp column studies.

3.4.2. Nikolaidis modified model

The experimental bed volume dependent breakthrough curves and model simulations based on Nikolaidis modified model for As(III) and As(V) are presented in Fig. 5. The values of the parameters are presented in Table 5.

This model also simulated the column experimental data reasonably well. The maximum adsorption capacities (q_m) estimated from the model for fresh IOCSp and regenerated IOCSp were 12.27 mg/g and 11.48 mg/g respectively. This value was nearly the same as the value calculated by Thomas model.

The equilibrium partitioning coefficient (K_d) shows the degree of arsenic adsorption or co-precipitation onto the IOCSp or surface binding. This values were 0.8L/mg for both As(III) and As(V). The K_d values in this study were lower than that of arsenic adsorption or co-precipitation onto the iron filing or surface binding (4.3 L/mg) [21]. The first-order mass-loss coefficient (k_N) shows the rate of mass of arsenic lost during solute arsenic transport to the surface of IOCSp. The values of k_N for both As(III) and As(V) were $7 \times 10^{-5}/s$.

Table 6 presents the breakthrough times for fresh IOCSp and regenerated IOCSp calculated from the Nikolaidis modified model and obtained experimentally.

Table 4

Comparison of the theoretical service times from the Thomas approach with the experimental time.

Species	Breakthrough time of fresh IOCSp (h)		Breakthrough time of regenerated IOCSp (h)	
	Experimental	Theoretical	Experimental	Theoretical
As(III)	580	584	467	480
As(V)	673	647	551	519

Table 5

Parameters of Nikolaidis modified model.

No	Parameters	Unit	Fresh IOCSp		Regenerated IOCSp	
			As(III)	As(V)	As(III)	As(V)
1	q_m	mg/g	12.27	12.27	11.48	11.48
2	k_N	1/s	7×10^{-5}	7×10^{-5}	7×10^{-5}	7×10^{-5}
3	K_d	L/mg	0.8	0.8	0.8	0.8
4	D_x	m ² /s	3×10^{-6}	1.9×10^{-6}	4×10^{-6}	2.5×10^{-6}
5	r		0.987	0.991	0.983	0.980

Table 6

Comparison of the theoretical service times from the Nikolaidis modified model with the experimental time.

Species	Breakthrough time of fresh IOCSp (h)		Breakthrough time of regenerated IOCSp (h)	
	Experimental	Theoretical	Experimental	Theoretical
As(III)	580	593	467	482
As(V)	673	675	551	572

As is evident from Table 6, the theoretical and experimental values were well comparable with each other. In addition, the correlation coefficients (r) values of this model were more than 0.98, representing a good correlation of the simulated data with observed data. These models are relatively simple and simulate the adsorption process of arsenic on IOCSp reasonably well. The Nikolaidis model was initially used for arsenic removal by zero valent iron but in this study, IOCSp, a very high porosity material which a specific surface area was used. Therefore, the accuracy of the model could be improved further by incorporating these factors.

4. Conclusions

IOCSp can adsorb both As(III) and As(V). The As(III) and As(V) adsorption capacities calculated by Sips isotherm were 4.2 mg/g and 4.6 mg/g, respectively. The IOCSp filter was able to be operated for a long period with consistent arsenic removal efficiency. A column (4.5 cm diameter and 78 cm IOCSp bed depth) packed with 25 g of IOCSp could maintain about 95% arsenic removal from high contaminated water containing arsenic of 1000 μ g/L for more than 153 L and 178 L of As(III) and As(V) respectively. IOCSp could be regenerated with NaOH and it did not have any significant decrease in removal as compared to the fresh IOCSp. The dynamic adsorption kinetics of arsenic on IOCSp can successfully be described and predicted by the Thomas and Nikolaidis modified models.

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