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Original Research Paper

Arsenic removal from water by physico-chemical approach – A Low-cost treatment option

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Methods available for arsenic removal from water are costly and cannot be applicable at large scale in the field. In the present study a low cost treatment of arsenic has been attempted which would be feasible in terms of village level applications. A laboratory investigation was carried out to study the removal of arsenic from water samples containing arsenic of known concentration by adsorption. Iron oxide coated and uncoated river sand of 0.50 mm geometric mean size has been used as adsorbent. Studies were carried out for finding the effects of various parameters like initial arsenic concentration, pH of arsenic solution, dose of adsorbent, contact time and filtration rates. Arsenic removal of 94% by coated sand and 40% by uncoated sand has been observed at optimum dose and initial concentration 1 mg/l of arsenic.

Key words: Arsenic removal, adsorption, low-cost treatment.

INTRODUCTION

Arsenic is a naturally occurring element found in soils and groundwater. Arsenic usually exists in two different forms, or valences, in a natural setting depending on the amount of oxygen available in groundwater. Arsenic (As) contamination of drinking water is a major health concern, because drinking arsenic contaminated water is linked to several types of cancers (WHO 1981). Unfortunately, there is no known cure for arsenic poisoning and therefore providing arsenic free drinking water is the only way to weaken the adverse health effects of arsenic. In more shallow aquifers with higher levels of oxygen, arsenic will usually exist as arsenate, As (V). In deeper, anaerobic ground waters, arsenic usually occurs as arsenite, As (III). When arsenic levels are found to be too high at a specific location, it may be necessary to treat drinking water to remove it (WHO 1984a).

It is important to note that the most effective way to overcome the adverse health effects of arsenic is prevention of further exposure by providing safe drinking water, because there is no effective treatment to counteract arsenic toxicity. Therefore, the World Health Organization (WHO, 1984) has recommended a maximum contaminant level (MCL) for drinking waters of 0.01 mg/L. Many countries however, permit higher arsenic concentrations in drinking water mainly due to the high cost of treatment to lower concentrations. As noted by Gregor (2001) the MCL value is the concentration below which the presence of arsenic is not considered to pose a significant health risk,even after a lifetime consumption of the water.

Both arsenite and arsenate have a high affinity for Fe-oxides (Goldberg and Johnston,2001; Smedley and Kinniburg, 2002), but the cost of the adsorptive metal removal process is high when pure sorbents (activated carbon) are used (Apak et al., 1998). The present study of removal of arsenic from water by iron-oxide coated sand was under taken with a view to provide low cost solution to the drinking water having arsenic problem.

The Practical Considerations

Contamination of groundwater with arsenic

High arsenic concentrations have been detected in groundwaters in several parts of the world. In India the most significant area of groundwater arsenic contamination in terms of the number of people affected, is West Bengal (Chakraborty and Saha 1987). The most important routes of arsenic exposure are through food and drinking water.

Arsenic is found in food, particularly in fish and shellfish, in which it is found mainly in the less toxic organic form (Vukašinović-Pešić et. al.,2005). Arsine is considered the most toxic form, followed by arsenites, then arsenates and organic arsenic compounds (Rajaković 1986, Wang et. al., 2004).Environmental exposure to arsenic through drinking-water has been associatedwith skin cancer (Zaldivar 1981). Acute arsenic intoxication associated with the ingestionof water containing a very high concentration of arsenic (21.0 mg/L) wasreported (WHO 2008). Moreover, it was concluded that long-term exposure to arsenic in drinkingwater is causally related to increased risks of cancer in the skin, lungs, bladderand kidney, as well as other skin changes, such as hyperkeratosis and pigmentationchanges (IPCS 2001). Epidemiological studies (Chen et. al., 1985) in areas with different frequencies of black-foot disease and where drinking water contained 0.35–1.14 mgL^{-1} arsenic revealed elevated risks for cancers of the bladder, kidney, skin, lung,liver and colon. Dermal lesions were the most commonly observed symptom occurring after minimum exposure periods of approximately 5 years (Chen et. al., 1986). High arsenic level in ground waters have been reported from many parts of the world, e.g., 0.6-2.0 mg/l in Taiwan (Kuo,1968, Shen 1973), 0.9-3.4 mg/l in Argentina (Arguello et al., 1938) and 0.07-1.7 mg/l in USA (Goldblatt et al., 1963). In India Datta and Kaul (1976) reported that 29.4 % of water samples from wells, 50% from tubewells and 35 % from springs in and around Chandigarh to contain high levels of arsenic. Chakraborty and Saha (1987) reported 0.64 mg/l mean arsenic level in tubewell (110-150 feet deep) water samples collected from several districts of West-Bengal, the rang being 0.2-2.0 mg/l.

Chemistry of arsenic

Arsenic (atomic wt. =74.9216) is a non-metal in group (a) of the periodic chart. The physical appearance of arsenic resembles that of a metal, so it is referred to as a metalloid to distinguish it from a true non-metal. It exists in several different oxidation states +V(arsenate), +III(arsenite), 0(arsenic)and –III (arsine) (Mcbride 1971). But in general it is found as an anion with acid characteristics in only the trivalent and pentavalent forms.The soluble arsenite species are H_3AsO_3,$H_2AsO_3^-$and $HAsO_3^{-2}$. The soluble arsenate species are H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^-$, $HAsO_4^{-2}$ and AsO_4^{-3}.

Comparing arsenic removal technologies

Adsorption systems are becoming more popular, especially in small scale treatmentsystems such as at the household level (Thirunavukkarasu et al., 2003). This is important in countries like Bangladesh, where the water system is not centralised and individual households or small groups are served by their own well (Mamtaz andBache, 2001).

Adsorption is a mass transfer process, which refers to the accumulation of sorbates at the liquid/solid interface. One of the most common methods for arsenic removal from water is the precipitation of arsenic as calcium arsenates or ferric arsenates at an optimum pH range (Bothe and Brown, 1999). In these precipitation processes, dissolved ions in a solution form an insoluble solid via a chemical reaction e.g. naturally occurring dissolved ion forms a precipitate when it is exposed to air.

Although ion exchange resins can be used to remove arsenic from water over a wide pH range, different resins have different exchange capacities; e.g. conventional sulphate resins are particularly suited for arsenate removal, but uncharged arsenite cannot be removed (Johnston and Heijnen, 2001). In addition, another concern is that competing anions, especially sulfate, obstruct arsenic removal because most of the resins are more selective towards sulfate.

The associated costs of these methods include pH adjustment, operation andmaintenance costs, the cost of the sorbent itself, and safe handling of the spent sorbent (disposal or regeneration). The advantages of this method (Kartinen and Martin, 1995;Manning and Goldberg, 1997; Bajpai et al., 1999; Chen et al., 1999; Chwirka et al.,2000; O'Reilly et al., 2001; Korngold et al., 2001) can be summarised as:

1. User friendly at the household level and, cheaper.
2. The efficiency of the ion exchange process is less sensitive to the pH of the water.

Therefore, there is need to research on new methods of arsenic removal involving low cost techniques. One of such emerging methods is the use of iron oxide coated sand.

MATERIALS AND METHODS

In the present study both coated and un-coated river sand of 0.50 mm geometric mean size has been used as adsorbents on prepared arsenic samples. The adsorption studies along with the effect of certain parameters like initial arsenic concentration, initial pH, contact time, dose of adsorbent etc. on the behaviour of arsenic, adsorbed on the adsorbents used were performed in batch study as well as column study. Column used, was prepared by using 25.4 mm (1 inch) internal diameter PVC pipe of length 160 cm (Figure 1) (Chaudhuri and Joshi 1996).

Sample preparation

Arsenic sample was prepared with distilled water by dissolving 1.32 gm of arsenic trioxide (As_2O_3) in 25 ml of a 20 % (W/V) KOH solution. The resulting solution was neutralised with 20 % (V/V) $H_2SO_{4.}$ The concentration of resulting solution was 1000 ppm. Samples of required concentrations were prepared by diluting this sample to

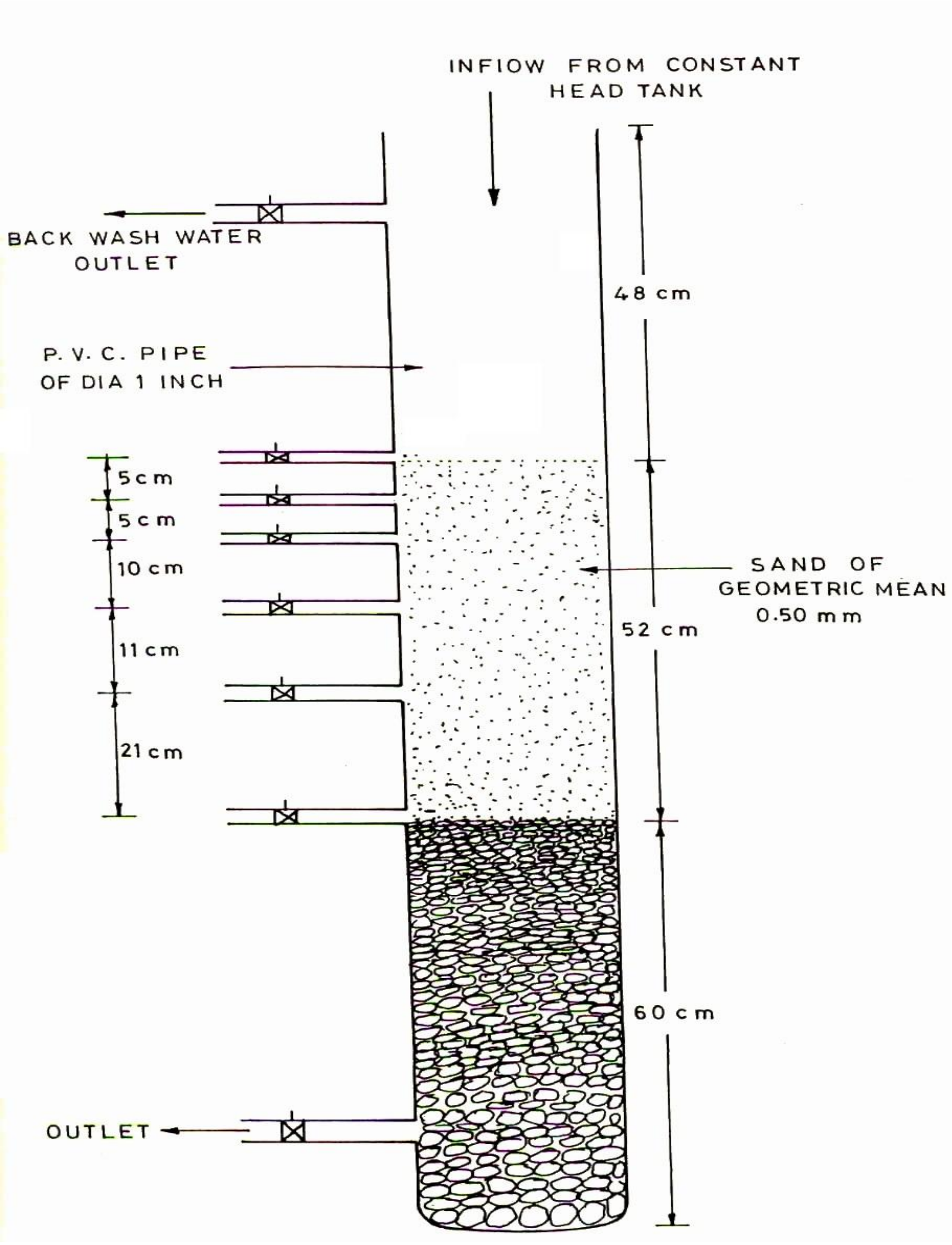

Figure 1: Filter Used for the Column Study

the required extent with distilled water.

Preparation of Iron oxide coated sand

Iron oxide–coated sand was prepared using a procedure similar to that of Bailey et al., (1992). For this purpose river sand was used. Washed and dried river sand (1800 gm) of geometric mean size 0.50 mm was mixed with 720 ml of 2.5 M ferric nitrate [$Fe(NO_3)_3.9H_2O$]solution(which was adjusted to pH 11.0 with sodium hydroxide) for two minutes. Then the mixture was placed at temperature of 110°C in drying oven for 24 hrs.

The coated sand was washed with distilled water until the runoff was clear, dried at 105°C and stored in capped

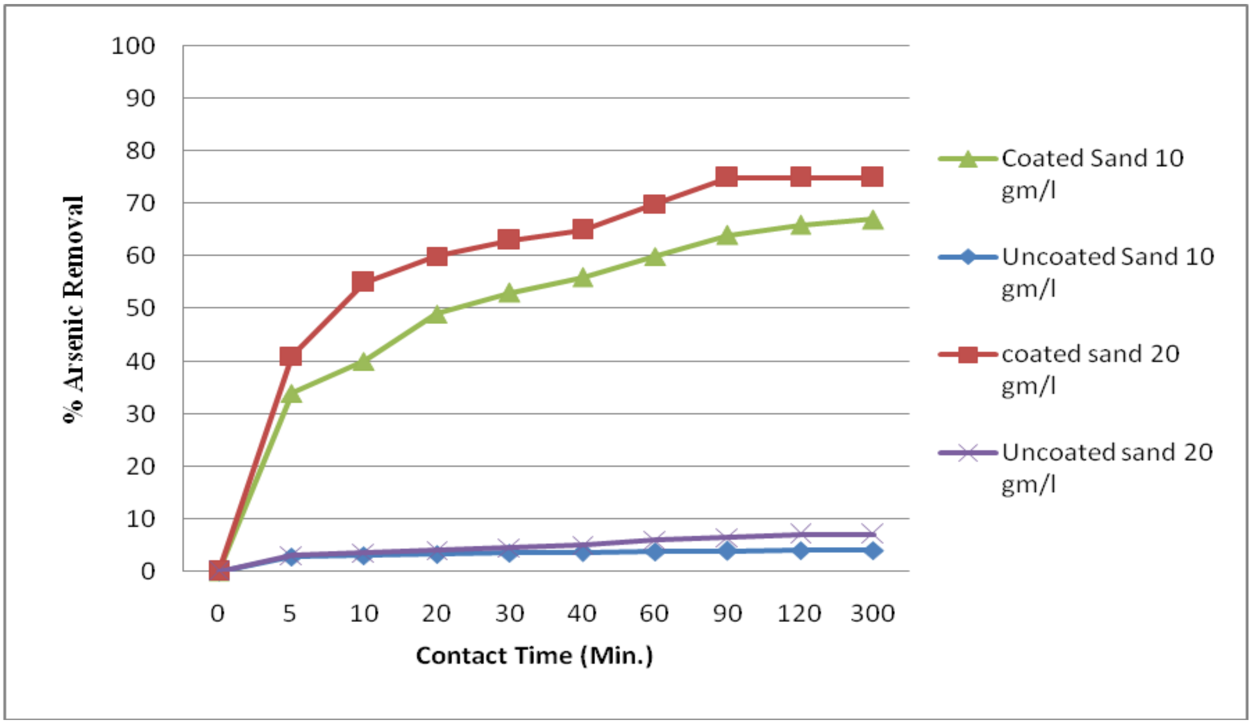

Figure 2: Effect of Contact Time on ARE

bottles.

Arsenic measurements

In the present study the arsenic is quantified by Inductively coupled plasma (ICP-MS). ICP analyses the arsenic at wave-length of 189.042 nm.

RESULTS AND DISCUSSION

The adsorption studies along with the effect of certain parameters like initial arsenic concentration, initial pH, contact time, dose of adsorbent etc. on the adsorption behaviour of arsenic have been performed in batch as well as column study. For these studies iron oxide coated and uncoated sand have been used as adsorbents.

Arsenic removal using sand in batch study

Effects of various parameters in batch studies are discussed below

Effect of contact time on ARE (Arsenic Removal Efficiency)

Initially adsorption increases rapidly with increasing contact time and after 2 hrs of contact time it becomes almost constant (Figure 2). It is also observed that with the same dose, time and initial arsenic concentration, coated sand removes higher % of arsenic as compare to uncoated sand. Results obtained are close to the results obtained by Choudhuri et al., (1996).Initially more surface is available for adsorption i.e. arsenic removal is faster but after a certain time adsorption and desorption becomes almost equal causing arsenic removal to be constant.

Effect of adsorbent dose on ARE

Effect of adsorbent dose on ARE has been analysed (Table 1). Initially adsorption efficiency for arsenic removal increases rapidly but after a certain dose it becomes almost uniform. At the dose of 40 gm/ml, 87% of arsenic removal was observed by coated sand while 12% of arsenic removal was obtained by uncoated sand. The results can be attributed to the fact that with increase in the adsorbent dose more and more adsorbent surface is available for solution to adsorb and this increases the % adsorption. However there is very slow increase in arsenic removal if we go beyond an optimum adsorbent dose because for adsorbent dose, adsorption sites are not easily accessible.

Effect of initial pH on ARE

Effect of initial pH on ARE has been analysed. It is observed that on increasing the pH of arsenic solution initially ARE increases rapidly up to a certain pH i.e. 10 and then becomes almost constant or reduces slightly. By coated

Table 1. Effect of Dose of Adsorbent on ARE

	Arsenic Removed (%) in Coated Sand		Arsenic Removed (%) in Uncoated Sand	
Dose gm/l	**Arsenic Concentration = 1 mg/l**	**Arsenic Concentration = 2.09 mg/l**	**Arsenic Concentration = 1 mg/l**	**Arsenic Concentration = 2.09 mg/l**
5	38%	29.9%	2%	2.12%
10	66%	68.9%	4%	4.30%
20	75%	75.6%	7%	8.13%
30	84%	84.7%	10%	12.15%
40	87%	87.6%	12%	15.78%
50	89%	89.8%	19%	19.60%

Table 2. Effect of Concentration of Arsenic on ARE

Arsenic Concentration mg/l	Arsenic Removed (%) in Coated Sand	Arsenic Removed (%) In Uncoated Sand
0.5	76	10
1	76	9
2	75	8
3	74	7
4	72	6
5	70	5
6	68	5

sand at I ppm initial arsenic concentration the % removal was observed as 75% at pH=10 and it decreases slightly to 74% at pH=11 for 20 gm/l dose.

In any adsorbate-adsorbent system pH affects the adsorbate and adsorbent nature and thus affects the extent and rate of adsorption. The decrease in the adsorption of arsenic at pH more than 10 may be due to the change in ionic form of arsenic (v).

Effect of concentration of arsenic on ARE

Effect of initial concentration of arsenic on ARE has been analysed (Table 2). Initially the arsenic adsorption efficiency was almost independent of initial arsenic concentration. At 0.5 mg/l initial arsenic concentration the % arsenic removal was 76% and at 6 mg/l concentration it is 68% for coated sand. Similarly for uncoated sand at 0.5 mg/l initial arsenic concentration the % arsenic removal was 10% while at 6 mg/l concentration it is 5%. Hence it can be concluded that at the lower concentration the % arsenic removal is almost independent of initial concentration but slight decrease in % of arsenic adsorbed was observed at higher initial concentrations. Results obtained are close to the results obtained in the adsorption study of arsenic by Janet et al., (1997).

From the batch studies it is concluded that at the same dose, contact time, initial arsenic concentration and pH, coated sand removes higher % of arsenic than uncoated sand. This may be due the reason that surface charge of sand could be made positive by adsorption of Fe^{+3} ions, and formation of ferric-arsenate complexes take place by these Fe^{+3} ions on the surface of the adsorbent. The total free energy of adsorption increases with increase in the charge of adsorbent due to increase in the energy of solvation interactions. Hence iron treatment does change the surface characteristics of sand and hence increase the adsorption capacity of sand or increase the % removal of arsenic.

Adsorption isotherms

Adsorption equilibrium data are conveniently represented by adsorption isotherms. Various types' adsorption isotherms i.e. Langmuir and Freundlich have been tried, but adsorbate-adsorbent combination Langmuir isotherm fits well. It might be possible that assumptions made in Langmuir isotherm are satisfied in these experiments.

The analysis of datafor the adsorption of arsenic on coated and uncoated sand has been done in the light of rearranged Langmuir isotherm model.

$$\frac{1}{C_e} + b = \frac{ab}{q_e}$$

Where C_e and q_e are adsorbate concentration in the solution and solid phase respectively and a, b are Langmuir constants. In order to examine the equilibrium behaviour plots between $\frac{1}{C_e}$ and $\frac{1}{q_e}$is drawn for both adsorbents at two different concentrations (Figure 3). The linearity of plots indicates compliances with the Langmuir isotherm.

Langmuir constants are evaluated from these plots and are

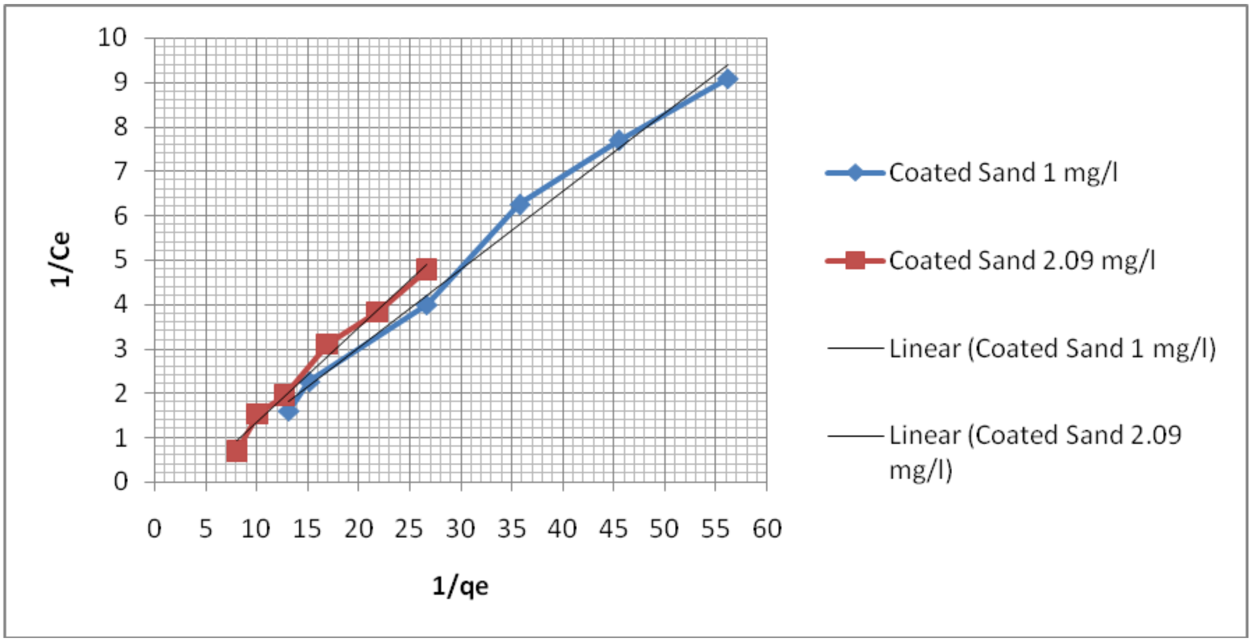

Figure 3: Langmuir Plot for the Adsorption of Arsenic on Coated Sand

Table 3. Langmuir Constants

Initial Concentration mg/l	Coated Sand			Uncoated Sand		
	Langmuir Constant (a) mg/gm	Langmuir Constant (b) 1/mg	$R_L=1/1+bC_0$	Langmuir Constant (a) mg/gm	Langmuir Constant (b) 1/mg	$R_L=1/1+bC_0$
1	0.253	0.67	0.598	0.011	0.231	0.812
2.09	0.198	1.11	0.301	0.336	0.027	0.946

reported in Table (3).

The essential characteristics of the Langmuir isotherm may be expressed in terms of a dimensionless equilibrium parameter R_L using following equation

$$R_L = \frac{1}{1+bC_0}$$

Where C_0 = Initial arsenic concentration

The values of RL were found between zero and one showing favourable adsorption. These are also reported in Table 3.

Using Coated Sand:

At initial arsenic concentration= 1 mg/l

$$\frac{1}{C_e} + 0.67 = \frac{0.17}{q_e}$$

At initial arsenic concentration= 2.09 mg/l

$$\frac{1}{C_e} + 1.11 = \frac{0.22}{q_e}$$

Using Uncoated Sand:

At initial arsenic concentration= 1 mg/l

$$\frac{1}{C_e} + 0.231 = \frac{2.5x10^{-03}}{q_e}$$

At initial arsenic concentration= 2.09 mg/l

$$\frac{1}{C_e} + 0.027 = \frac{9.09x10^{-03}}{q_e}$$

The values of Langmuir constants obtained are close to the values obtained by Choudhuri et al., (1996).

Arsenic removal kinetics

The % removal and contact time are drawn on log-log scale and curves obtained are straight lines.Following are the equations for different straight lines:

at1 ppm arsenic concentration, using coated sand

For the dose of 10 gm/l

$p=31.5\ t^{0.2}$

For the dose of 20 gm/l

$p=33.45\ t^{0.33}$

at 1 ppm arsenic concentration, using uncoated sand

For the dose of 10 gm/l

$p=2.72\ t^{0.1}$

For the dose of 20 gm/l

$p=2.58\ t^{0.25}$

at 2.09 ppm arsenic concentration, using coated sand

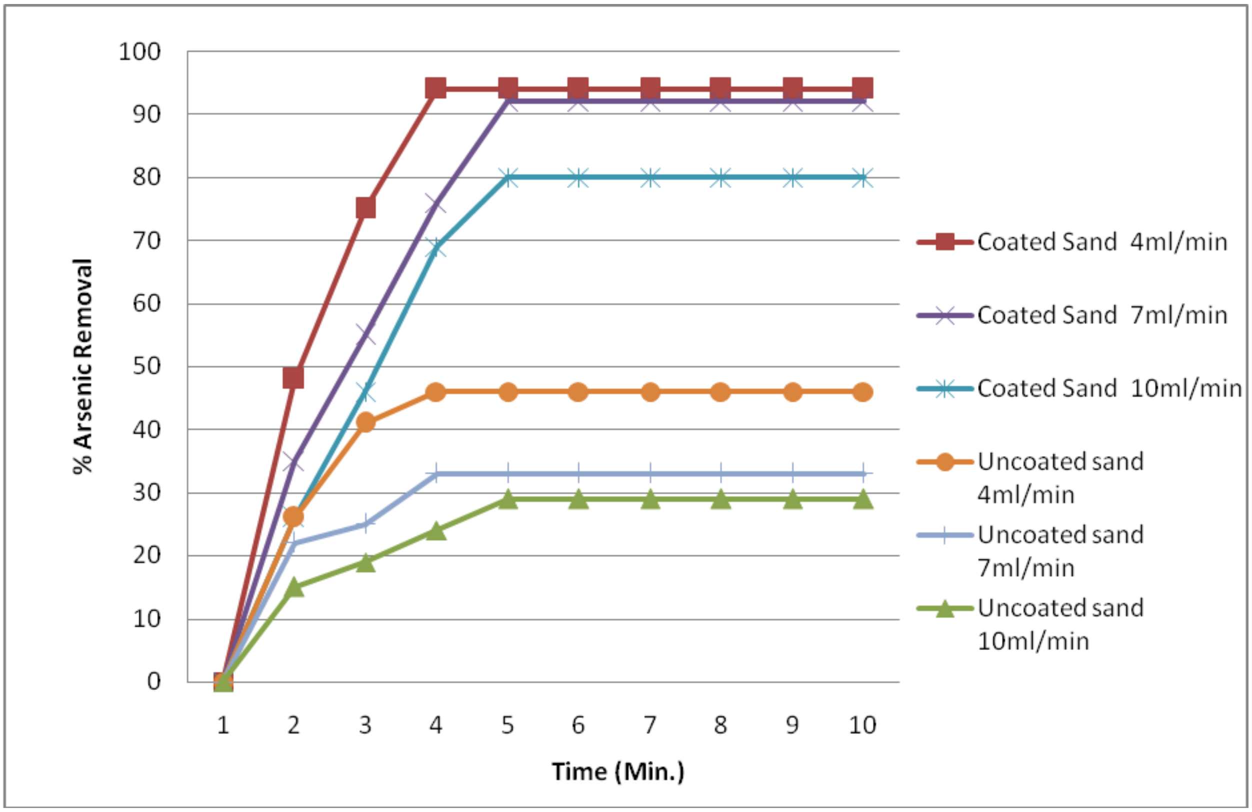

Figure 4: Effects of Filtration Rates on ARE

For the dose of 10 gm/l

$p=1.65\ t^{0.25}$

For the dose of 20 gm/l

$p=2.72\ t^{0.30}$

Where p=% arsenic removal and t= contact time in minutes

If the interparticle diffusion was the rate determining step, the rate of adsorption would have varied with the square root of the time producing the equation of line as $p=ct^{0.5.}$ The type of the adsorption observed in these experiments was not merely controlled by diffusion rate since removals were the functions of t0.1to $t^{0.33}$rather than $t^{0.5}$. Rate determining step may be the migration of arsenic particle from solution to adsorbent or boundary layer diffusion.

Effects of filtration rate on ARE

Effect of filtration rates on ARE has been shown in Figure 4. From it clear that with the increase of filtration rates, ARE decreases. Using coated sand the % of removal of arsenic after 3 hours at 52 cm bed depth was found as 94% at 4ml/min filtration rate while it was 80 % at 10 ml/min keeping the other conditions same.

At filtration rate 10 ml/min, if H- head loss in mm and t-time in minutes

$H=0.607\ t^2+5.178t+0.857$

At filtration rate 7 ml/min

$H=0.261t^2+3.214t+0.238$

At filtration rate 4 ml/min

$H=0.492t^2-0.178t+0.014$

In case of uncoated sand for bed depth of 52 cm at 4ml/min the % removal of arsenic was 46% after 3 hours while it was 29% at the rate of 10 ml/min.

At filtration rate 10 ml/min

$H=0.726\ t^2+2.19t+1.381$

At filtration rate 7 ml/min

$H=0.202t^2+2.392t+0.190$

At filtration rate 4 ml/min

$H=0.309t^2-0.642t+0.047$

From the head loss pattern is clear that as the filtration rate increases, the head loss also increases with time. In coated sand after 6 hours of filtration time the head loss was found to be 54 mm at 10 ml/min filtration rate while it was 17 mm at the filtration rate of 4 ml/min. Similar pattern was obtained for uncoated sand but the values of the head loss were less than the corresponding values of head loss for coated sand.

Effects of bed depth of filter on ARE

Effect of bed depth of filter on ARE has been analysed. As the bed depth increases the % removal of arsenic also increases. At the 7 ml/min filtration rate the removal of arsenic was 91% at 52 cm bed depth while it was only 51% at 10 cm bed depth, after 3 hours for coated sand. Similarly for uncoated sand the % removal of arsenic was 40 % at 52 cm bed depth while it was only 16 % at 10 cm bed depth after 3 hours at 7 ml/min filtration rate. Reason for this behaviour can be explained on the basis of the fact that with the increase of bed depths, the more surface area will be available for adsorption and hence ARE increases.

Using coated sand, after 24 hours for 7 ml/min the head was found as 145 mm at 52 cm bed depth while it was 52 mm at 10 cm bed depth keeping all other conditions same.

At Bed Depth 52 cm

$H=0.111\ t^2+3.341t+0.436$

At Bed Depth 31 cm

$H=0.115t^2+1.759t+1.423$

At Bed Depth 20cm

$H=0.073t^2+0.430t+0.328$

At higher bed depths the adsorption rate of arsenic is more and hence porosity decreases rapidly and therefore head loss increases in bed depths.

Effect of initial pH on ARE in filtration

It was observed that the % removal of arsenic increases with increase in pH up to 10, after which it starts decreasing slowly on increasing the pH. For coated sand the maximum % removal of arsenic was 92% at pH= 10.

CONCLUSIONS

In the present work batch study and column study for adsorption of arsenic on iron-oxide coated and un-coated sand, were performed. The various variables investigated were contact time, adsorbent dose, pH, and initial arsenic concentration, bed depth of filter and filtration time. Following conclusions are drawn from the present study:

(1) Coating of iron oxide coated sand increases the arsenic removal efficiency of sand very effectively. Hence iron-oxide coated sand showed promise as medium for use in small or home-treatment units for removing arsenic from water.

(2) In alkaline pH range the arsenic removal efficiency is better than in acidic or normal pH range.

(3) After a certain contact time arsenic removal occurs at slower rate. Similarly after certain filtration time arsenic removal reaches to its saturation stage.

(4) The extent of arsenic adsorption was relatively independent of initial arsenic concentration. A slight decrease in % of arsenic adsorbed occurs at higher concentrations.

(5) On increasing the adsorbent dose, arsenic removal increases and it becomes almost steady after a certain optimum dose.

(6) Equilibrium experimental data confirms well to the Langmuir isotherms for both the adsorbents.

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