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Effects of adsorbent dose, its particle size and initial arsenic concentration on the removal of arsenic, iron and manganese from simulated ground water by Fe³⁺ impregnated activated carbon

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Abstract

This paper presents the observations of the study on arsenic removal from a contaminated ground water (simulated) by adsorption onto Fe³⁺ impregnated granular activated carbon (GAC-Fe). Fe²⁺, Fe³⁺ and Mn²⁺ have also been considered along with arsenic species in the water sample. Similar study has also been done with untreated granular activated carbon (GAC) for comparison. The effects of adsorbent dose, particle size of adsorbent and initial arsenic concentration on the removal of As(T), As(III), As(V), Fe²⁺, Fe³⁺ and Mn²⁺ have been discussed. Under the experimental conditions, the optimum adsorbent doses for GAC-Fe and GAC have been found to be 8 g/l and 24 g/l, respectively with an agitation time of 15 h. Particle size of the adsorbents (both GAC and GAC-Fe) has shown negligible effect on the removal of arsenic and Fe species. However, for Mn removal the effect of adsorbent particle size is comparatively more. Percentage removal of As(T), As(V) and As(III) increase with the decrease in initial arsenic concentration (As₀). However, the increase in percentage removal of all the arsenic species with decrease in As₀ are less for higher value of As₀ (3000–500 ppb) than those of the lower value of As₀ (500–10 ppb). The % removal of As(T), As(III), As(V), Fe, and Mn were ~95%, 92.4%, 97.6%, 99% and 41.2%, respectively when 8 g/l GAC-Fe was used at the As₀ value of 200 ppb. However, for GAC these values were ~55.5%, 44%, 71%, 98% and 97%. The pH and temperature of the study were 7 ± 0.1 and 30 ± 1 °C, respectively. © 2007 Elsevier B.V. All rights reserved.

Keywords: Arsenic; GAC; Iron impregnated activated carbon; Surface modified adsorbent; Manganese; Adsorption

1. Introduction

Arsenic, the hazardous chemical most widely happened in the world [1], is found in the shallow zones of ground water of many countries like Argentina, Bangladesh, Cambodia, Canada, Chile, China, Germany, Hungary, India, Mexico, Mongolia, Myanmar, Nepal, Pakistan, Romania, Thailand, USA, Vietnam, etc. in various concentrations. In some places in Bangladesh its concentration is as high as 1000 µg/l [2].

Arsenic contamination in water has posed severe health problems around the world. Considering the lethal impact of arsenic on human health, environmental authorities have taken a more stringent attitude towards the maximum contaminant level (MCL) of arsenic in water. World Health Organization (WHO) in 1993 and National Health and Medical Research Commit-

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tee (NHMRC), Australia, in 1996 had recommended MCL of arsenic in drinking water as 10 and 7 μ g/l, respectively [3]. The MCL of arsenic in drinking water has also been reduced from 50 to 10 μ g/l by European Commission in 2003 [4]. Environmental Protection Agency (EPA), USA, has decided to move forward in implementing the same MCL of arsenic that is recommended by WHO for drinking water in 1993 [5]. Japan and Canada have reduced the MCL for arsenic in drinking water to 10 and 25 μ g/l, respectively. The MCL for arsenic in countries like India, Bangladesh, Taiwan, China, Vietnam, etc. is also 50 μ g/l [6].

In recent years, use of surface modified adsorbents for the development of cheaper arsenic removal technique has acquired momentum. Recently, some adsorbents like Cu impregnated coconut husk carbon, iron oxide coated polymeric materials, iron oxide coated sand, iron oxide coated cement, bead cellulose loaded with iron oxy hydroxide, etc. have been reported [2] for effective adsorption. Relatively very little information is published on the adsorption of arsenic species by activated

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carbon. However, the use of iron impregnated granular activated carbon (GAC-Fe) has been reported by some researches [7–11] for removing arsenic from water. By the impregnation of Fe^{3+} onto the surface of untreated granular activated carbon (GAC) an amorphous layer of FeOOH is formed as per the following equation [12]:

$$Fe^{3+} + 3OH^- \rightarrow FeOOH + H_2O$$

Formation of such layer of FeOOH increases the net positive charge (NPC) of the GAC-Fe [9,13] and improves the arsenic adsorption capacity. This iron in GAC-Fe activates the oxidation of As(III) to As(V), which can easily be adsorbed by the adsorbent in the experimental pH range [14]. In case of GAC, As(III) does not oxidize to As(V). However, some amount of As(III) may be adsorbed as per the following equation for both GAC and GAC-Fe [15]:

$$Fe(OH)_{(s)} + H_3AsO_{3(aq)} \rightarrow FeHAsO_{3(s)}^{2-} + H_2O$$

In literature GAC-Fe dose has been considered in a wide range (0.2-80 g/l) for the removal of arsenic from contaminated water [7–11]. However, the comparative removal of total arsenic (As(T)), As(III) and As(V) are not reported in these studies. The effects of adsorbent dose, adsorbent particle size and initial arsenic concentration on the removal of arsenic species have not been reported in detail. The comparison between the adsorption efficiency of the GAC and iron impregnated GAC is also hardly reported. The effect of other interfering metal ions, which are frequently present in contaminated ground water, on the removal of arsenic species has also rarely been investigated so far.

This paper explores the possibilities of the ferric chloride impregnated GAC to remove arsenic species in presence of iron and manganese ions which are available frequently in ground water. The effect of adsorbent dose, its particle size and initial arsenic concentration on the removal of arsenic species along with Fe and Mn have been reported. The adsorption capacity of GAC and iron impregnated GAC for the removal of the Fe and Mn have also been compared.

2. Materials and methods

Properties of GAC and GAC-Fe

All the chemicals, purchased from S.D. Fine-Chem Limited, India, were of reagent grade and solutions were prepared by Milli-Q water (Q-H₂O, Millipore Corp. with resistivity of 18.2 M Ω cm). The stock solutions of 100 ppm As(V) and As(III) were prepared by dissolving Na₂HAsO₄·7H₂O and NaAsO₂ in water and filtered through a 0.45 μ m membrane.

2.1. Preparation of GAC-Fe

Granular activated carbon of bulk density 40 g/100 ml was ground and sieved to various fractions of particle range 0.125-0.150 mm, 0.710-0.850 mm, 1-2 mm, 2-4 mm and 4-5 mm with the help of standard sieves. All the ground materials of various particle sizes were washed by Millipore water and dried at 105 °C till the constant weight was observed. Hundred grams of the dried material of each fraction was treated with 240 ml of ferric chloride solution containing 2.5% Fe³⁺ $(pH \cong 6.8 \pm 0.2)$, the pH was raised to 12 by the addition of 1N NaOH solution. The optimum value of Fe³⁺ was taken as per the finding of Gu et al. [9]. The high pH value for impregnation was considered for generation of maximum surface charges on the surface of GAC-Fe. The impregnation was carried out at 70 °C on a water bath till the complete evaporation of water was observed and then it was dried at 110 °C for 24 h [16]. The dried material was washed with Millipore water till the washing liquid became free from iron and then dried to constant weight. No colour change of wash liquid after the addition of silver nitrate and KSCN solution indicated absence of chloride and iron in the wash liquid, respectively. The iron content of the dried GAC-Fe was \sim 4.78%. Fe was taken into liquid phase from GAC-Fe by leaching with strong HNO₃ and was measured by AAS, GBC, Avanta, Australia. The changes in the key properties of the GAC due to Fe^{3+} impregnation are shown in Table 1.

Bulk density was measured by a picnometer, elemental analysis of the GAC and GAC-Fe was carried out by an elemental analyzer system (Elementar Analysensysteme GmbH, model Vario-EL V3.00). Surface area and micro pore volume of the samples were measured by N2 adsorption isotherm using an ASAP 2010 Micromeritics instrument by Brunauer-Emmett-Teller (BET) method, using the software of Micromeritics. Nitrogen was used as cold bath (77.15 K). SEM photograph was taken by an electron microscope (LEO Electron Microscopy Ltd., England). X-ray diffraction pattern was taken from a Cu target X-ray diffractometer (model D8 Advance, BRUKER aXS) with $2\theta = 5-100^{\circ}$. IR spectra of the adsorbents have been taken by a Thermo FTIR (model AVATR 370 csl) coupled with EZOMNIC software (version 6.2). Around 10 mg of dried sample was dispersed in 100 mg of spectroscopic grade KBr to record spectra.

2.2. Procedure

For each experiment 50 ml of the synthetic water sample containing 200 ppb As(As(III):As(V) = 1:1), 2.8 ppm Fe

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Adsorbent	Particle size (mm)	Elemental analysis (%)	Proximate analysis (%)	BET Surface area (m ² /g)	Micropore volume (cm ³ /g)	Bulk density (g/l)	
GAC	0.125-0.150	C: 75.06, H: 1.90, N: 0.0, S: 0.0, others: 23.04	Ash: 2.58, moisture: 9.71, others: 87.71	949.32	0.3316	756.27	
GAC-Fe	0.125-0.150	C: 74.39, H: 1.57, N: 0.30, S: 0.15, others: 23.59	Ash: 2.93, moisture: 8.85, others: 88.22	771.24	0.2746	822.51	

Table 2Operating parameters with their range

Operating parameters	Range
Temperature °C	30 ± 1
pH	7 ± 0.1
Adsorbent type and dose (g/l)	0-40 for both GAC and GAC-Fe
Adsorbent particle size (mm)	0.125-0.150 to 4-5
As ₀ (ppb)	0–3200

 $(Fe^{2+}:Fe^{3+}=1:1)$ and 0.6 ppm Mn was added with calculated amount of adsorbent in 100 ml vessel [17]. The sample was agitated in a shaker incubator for 15 h at 30 °C with shaking speed of 180 rpm [7]. To study the effect of adsorbent dose (AD) on the removal of arsenic species, the adsorbent dose was varied from 4 to 40 g/l. The initial pH of the solution was 7.1. It was measured after every 2 h interval and maintained at 7.1 ± 0.1 by the drop wise addition of N/10 HNO₃ when required. Particle size of the adsorbent (P.S.A.) was 0.125-0.150 mm. To investigate the effects of initial arsenic concentration and adsorbent particle size on the removal of arsenic, Fe and Mn the dose of GAC and GAC-Fe were 24 g/l and 8 g/l, respectively. After each experiment, the solution was filtered through 0.45 µm membrane filter. The filtrate was analyzed for total arsenic by a Perkin-Elmer ICP-MS (model ELAN-DRC-e). Arsenic speciation was done by Edward's ion exchange method [18]. The strong base anion resin AG 1 X8 was procured from Bio Rad. The analysis of iron and Mn was done by atomic absorption spectroscopy AAS, GBC, Avanta, Australia. All the experiments were repeated thrice and average results have been reported. The range of operating parameters is given in Table 2.

3. Results and discussions

Removal of arsenic species, iron and manganese by GAC and GAC-Fe is discussed in the subsequent sections.

3.1. Effect of adsorbent dose on the removal of arsenic

The removal of As(T), As(III) and As(V) by GAC and iron impregnated GAC (GAC-Fe) are shown in Fig. 1. From this

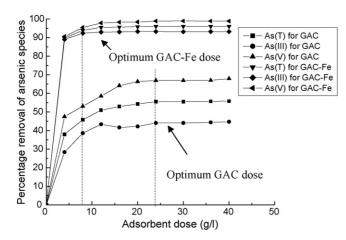


Fig. 1. Effect of adsorbent dose on the percentage removal of arsenic.

figure it is evident that at a constant As₀ value, the percentage removal of all arsenic species increase with the increase in adsorbent dose for both GAC and GAC-Fe. At lower adsorbent dose, for both GAC and GAC-Fe, the increase in percentage removal of arsenic species due to the increase in adsorbent does is very high. However, at higher adsorbent dose the increase in percentage removal of arsenic species due to the increase in adsorbent does is negligible. Beyond the adsorbent dose of 24 g/l and 8 g/l for GAC and GAC-Fe, respectively, the increase in the % removal of arsenic with increase in adsorbent dose is very less. Hence, the optimum removal of arsenic species can be obtained by using 8 g/l GAC-Fe and 24 g/l GAC. For both GAC-Fe and GAC this optimum removal is ~98.5% of the removal obtained at equilibrium after 72 h of agitation [10]. At the optimum dose the % removal of As(T), As(III), As(V) are $\sim 95\%$, 92.4% and 97.6%, respectively when GAC-Fe is used. For GAC these values are \sim 55.5%, 44% and 67%. Similarly, the specific uptakes for the adsorption of As(T), As(III) and As(V) by GAC-Fe, under the experimental conditions are $23.75 \,\mu g/g$, $23.1 \,\mu g/g$ and 24.8 µg/g, respectively. Whereas, for GAC these values are $4.63 \,\mu g/g$, $3.39 \,\mu g/g$ and $5.86 \,\mu g/g$, respectively. These observations indicate that the capacity of GAC-Fe for the removal of As(III), As(V) and As(T) are around seven, four and five times more respectively than that of GAC.

With the increase in adsorbent dose the number of active sites in unit volume of solution increases, which leads to the increase in the % removal of arsenic. However, after the adsorbent dose of 8 g/l this increase in % removal with the increase in GAC-Fe dose is less. It may be due to the two stages of arsenic adsorption (i.e., fast formation of mono layer followed by slow plateau stage) [19]. Recently, the % removal of As(T), As(III) and As(V) due to the adsorption on GAC-Fe with an adsorbent dose of 30 g/l and a shaking time of 8 h have been reported as ~95.5%, 93% and 98%, respectively [11]. A large number of GAC-Fe materials have also been reported recently in literature for the removal of arsenic species from water [7–9,11]. These GAC-Fe materials also achieve high arsenic removal over a range of water quality and adsorbent dose (3–35 g/l) conditions.

Under the experimental conditions the percentage removal of As(V) is 72% more than that of As(III) when GAC is used. However, for GAC-Fe the % removal of As(V) is only 5.6% more than that of As(III). This indicates that the GAC-Fe equally adsorbs As(III) and As(V) where as GAC adsorbs As(V) preferentially. For GAC the arsenic removal is possible due to the presence of positively charged active sites, which are developed due to the presence of free ash and metal oxides like Al₂O₃, CaO, SiO₂, etc. [20]. The SEM of GAC before and after adsorption as shown in Fig. 2a and b, also support the adsorption of arsenic onto the active sites of the GAC surface. At neutral pH range, As(III) exists as neutral species. Hence, its adsorption is less in the experimental conditions (pH 7 ± 0.1) [21]. Comparing the SEM of GAC and GAC-Fe before adsorption (Fig. 2a and c) it is also evident that an amorphous layer is formed on GAC due to the impregnation of Fe³⁺. Due to this reason the specific surface area of GAC-Fe is less than GAC-Fe (Table 1). Similarly, more adsorption on GAC-Fe is evident by comparing the SEM of GAC and GAC-Fe after adsorption (Fig. 2a and d). The extra

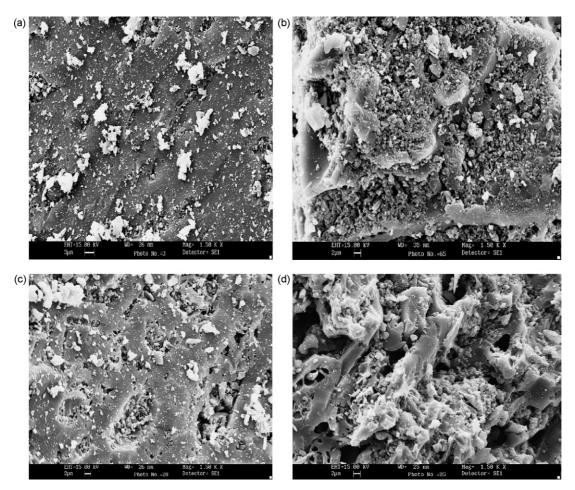


Fig. 2. SEM of GAC and GAC-Fe: (a) before adsorption of 0.00463 mg As/g GAC at a magnification of 1.5 k, (b) after adsorption of 0.00463 mg As/g GAC at a magnification of 1.5 k, (c) before adsorption of 0.02375 mg As/g GAC-Fe at a magnification of 1.5 k, and (d) after adsorption of 0.02375 mg As/g GAC-Fe at a magnification of 1.5 k, particle size of each was 0.125–0.150 mm.

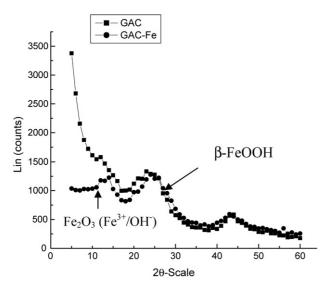


Fig. 3. X-ray diffraction pattern of GAC and GAC-Fe.

peak on the XRD of GAC-Fe at $2\theta = 12^{\circ}$ and higher peak at $2\theta = 28^{\circ}$ in Fig. 3 indicates the presence of Fe₂O₃ (Fe³⁺/OH⁻) and β -FeOOH onto the GAC-Fe, respectively [22,23], which are produced during the impregnation. Due to the formation of such layer, As(III) is partially converted to As(V) as a result GAC-Fe equally adsorbs As(III) and As(V) in contrast to GAC.

It is also evident that As(III) removal reaches its optimum value at a slightly lower dose of GAC-Fe than that of As(V). From this observation it seems that As(III) is transferred to As(V) before adsorption and the adsorption of As(V) is slower than that of conversion of As(III) to As(V) within the experimental conditions. It is also possible if some amount of As(III) is adsorbed directly. The more peak area at λ value of \sim 860 cm⁻¹ than that at λ value of $\sim 780 \,\mathrm{cm}^{-1}$ in the FTIR spectra of GAC-Fe after adsorption (Fig. 4) supports the partial conversion of As(III) to As(V). Similar observation on the As(III) and As(V) adsorption by amorphous iron oxide was reported recently [21]. The addition of bands at the wave number of \sim 825, and \sim 860 cm⁻¹ in the spectrum of GAC-Fe after adsorption indicates the adsorption of As(V) onto the GAC-Fe [24]. Again, the band at \sim 780 cm⁻¹ in the spectrum of GAC-Fe after adsorption indicates the direct adsorption of As(III) onto the GAC-Fe.

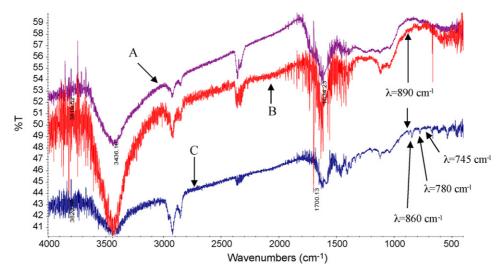


Fig. 4. FTIR spectra of GAC and GAC-Fe: GAC before adsorption (A), GAC-Fe before adsorption (B) and GAC-Fe after adsorption (0.02375 mg As/g GAC-Fe) (C).

Therefore, the As (III) removal capacity of GAC is improved due to the formation of GAC-Fe. From Fig. 4 it is also evident that for GAC-Fe the band area of the FTIR spectra (spectra B) at the wave number of \sim 3400 cm⁻¹ is reduced after adsorption of arsenic on GAC-Fe surface (spectra C). The spectral band at this wave number is for the stretching of Fe–OH bond. Therefore, the decrease in stretching band due to adsorption of arsenic suggests the addition of As(V) on Fe ions by replacing OH in the Fe–OH [25]. The more band area of spectra B at this wave number than that of spectra A also suggests the increase of FeOH bonds on GAC due to Fe³⁺ impregnation.

3.2. Effect of adsorbent dose on the removal of Fe and Mn

Fig. 5 shows the molar loading of Fe and Mn with varying adsorption dose. It is evident from the graph that around 98% removal of Fe and Mn is possible by GAC at the adsorbent dose of 24 g/l. The corresponding molar loading of Fe

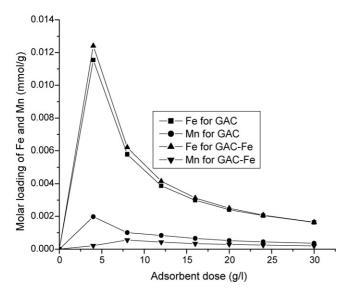


Fig. 5. Effect of adsorbent dose on the removal of Fe and Mn.

and Mn are 0.00205 mmol/g and 0.00044 mmol/g, respectively. For GAC-Fe at the adsorbent dose of 8 g/l the Fe removal is also 99% (molar loading is ~0.006207 mmol/g). However, for Mn the removal is only around 41% (molar loading is ~0.00056 mmol/g) at the adsorbent dose of 8 g/l. The Mn concentration in the treated water decreases with the increase in adsorbent dose of GAC-Fe. At an adsorbent dose of 8 g/l the percentage removal of Mn is 41% (molar loading is ~0.00056 mmol/g), which increases to 54% (molar loading is ~0.00025 mmol/g), when the GAC-Fe dose is 24 g/l. At neutral pH range Fe²⁺ may be oxidized to Fe³⁺ by the following equation:

$$Fe^{2+} + (1/2)O_2 + H^+ \rightarrow Fe^{3+} + (1/2)H_2O_2$$

This Fe³⁺ forms negatively charged moiety FeOO⁻, as a result \sim 99% removal of Fe²⁺ and Fe³⁺ is obtained for GAC-Fe. Some part of the Fe and Mn may also be removed due to surface precipitation in this pH range.

Mn cannot produce such negatively charged moiety in the neutral pH range. Again, with the impregnation of Fe³⁺ onto the surface of GAC the density of negatively charged sites on its surface is reduced at the neutral pH range. Due to this reason GAC can remove Mn ions completely at the adsorbent dose of 24 g/l but GAC-Fe can remove only 54% of Mn (molar loading is ~0.00025 mmol/g) with this adsorbent dose. Absorption peaks at 890 cm⁻¹ in spectra B and C indicate the presence of FeOOH [26]. Similarly absorption peak at 745 cm⁻¹ in spectra C indicates the adsorption of Mn on GAC-Fe [27]. The use of manganese for impregnation of GAC to improve the arsenic removal capacity has recently been reported [7]. However, they did not consider the Mn removal by this impregnated activated carbon, which normally occurs in ground water.

3.3. Effect of particle size on the removal of arsenic

The increase in percentage removal of all arsenic species with the decrease in particle size (from 4–5 mm to 0.125–0.150 mm)

of the adsorbent is very small (less than 2.0%). Among the particle sizes considered in the experiment the percentage removal is maximum for the particle size of 0.125-0.150 mm for both GAC and GAC-Fe. For GAC and most granular adsorbents the internal pore surface area is much bigger than the outer surface area. Hence, a reduction of the particle size (by grinding) does not lead to a higher total number of active sites available. However, the higher relative number of sites at the outer adsorbent surface results in more favourable kinetics. Therefore, the reduction of particle size does not improve the percentage removal much in these adsorbents. However, the lesser the particle size more is the grinding cost. Hence, the optimum particle size for both adsorbents may be considered as 2-4 mm. Particle size of this range has also been reported in literature for the removal of arsenic from contaminated water [17]. However, these reactions are time dependent because diffusion into the pores takes time.

3.4. Effect of particle size on the removal of Fe and Mn

For GAC all the particle sizes give more than 99% removals of Fe and Mn. For GAC-Fe the percentage removal of Fe is also around 99% for all the particle sizes. However, the percentage removal of Mn at the particle size of 4-5 mm is around 38% only which increases to around 41% at the particle size of 0.125-0.150 mm. It is important to note that the adsorption of Mn on the surface of GAC-Fe is more influenced by its particle size than the other elements. This indicates that the Mn adsorption occurs by physical adsorption (outer sphere complex) rather than chemical adsorption. Again, at the experimental pH range, Mn exists as positively charged moiety which is less adsorbed on the positive surface of the GAC-Fe by chemical attraction but physical adsorption may occur in considerable amount. By the decrease in the particle size the outer surface of GAC-Fe increase, hence more % removal of Mn is obtained with smaller particle size of GAC-Fe. However, these reactions are time dependent because diffusion into the pores takes time.

3.5. Effect of initial arsenic concentration on the removal of arsenic

Initial arsenic concentration (As₀) influences the percentage removal of arsenic species. At constant adsorbent dose (8 g/l for GAC-Fe and 24 g/l for GAC) the effect of As₀ on the percentage removal is shown in Fig. 6. It is evident that the percentage removal of all the arsenic species by the adsorption on GAC and GAC-Fe increases due to the decrease in As₀. It is interesting to note that for both GAC and GAC-Fe the slopes of the curves are steeper when the As₀ value lies within 0–500 ppb than when it is above 500 ppb. This indicates that the increase in percentage removal of both As(III) and As(V) with decrease in As₀ value is less for higher value of As₀ (3000–500 ppb) than that for lower value of As₀ (<500 ppb). This can be explained as follows.

It is a well-known fact that at a particular environment the percentage removal of an adsorption process depends upon the ratio of the number of adsorbate moiety to the available active sites of adsorbent. This ratio is also related to the surface coverage of the adsorbent (number of active sites occupied/number of active sites available) that increases with increase in the number of adsorbate moiety per unit volume of solution at a fixed dose of adsorbent. Less is the value of this ratio is high and decreases gradually with the decrease in As_0 as a result the % removal increases.

To explain more steepness of the curves in Fig. 6 at As_0 value <500 ppb, it can be hypothesized that most of the As(III) is first oxidized to As(V) in presence of GAC-Fe and is subsequently adsorbed onto the surface of the GAC-Fe. In case of GAC the oxidization of As(III) is less possible. However, the solution itself contains As(V) therefore the steepness of the curves in Fig. 6 for GAC is also possible. This As(V) is specifically adsorbed onto an oxide surface via ligand exchange mechanism and exists as an inner sphere surface complex [9].

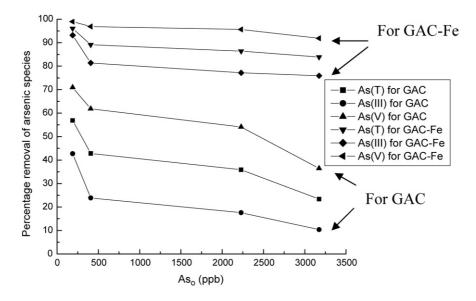


Fig. 6. Effect of initial arsenic concentration on the removal of arsenic species.

For adsorbents having low surface coverage with As(V), formation of mono dentate surface complex is considered to be predominant over bi-dentate complex [28]. It is possible that the formation of bi-dentate mono-nuclear complexes at high surface coverage is slower than the predominantly mono dentate reactions at low surface coverage [29]. It may be the fact that at an As₀ value of 500 ppb or above the bi-dentate mononuclear complexes are formed and below this As₀ value mono-dentate complexes are pre-dominant. As a result, the increase in % removal due to the decrease in As₀ value is more when As₀ value is less than 500 ppb, which gives steeper curves.

Using GAC-Fe the total arsenic content in the treated water may be reduced below 10 ppb if the As_0 value lies within 200 ppb. It is also evident that the As(T) in the treated water is below 50 ppb when As_0 value is 520 ppb or less. However, GAC can not reduce the As(T) below 50 ppb even if the As_0 value is 200 ppb. In India and Bangladesh the arsenic content in ground water varies from 50 to 300 ppb. Therefore, this GAC-Fe may be used to treat the contaminated ground water in this region.

3.6. Effect of initial arsenic concentration on the removal of Fe and Mn

The effect of As_0 is negligible for Fe and Mn removal by GAC. For GAC-Fe also Fe removal is independent of As_0 . However Mn removal is slightly increased with the increase in As_0 value. The reason for this slight increase of Mn removal with increasing As_0 is not so clear. However, this may be due to the less release of Mn by the GAC-Fe with the increase in As_0 .

4. Conclusions

From the above discussions the following conclusions are made:

- 1. The capacity of GAC-Fe for the removal of As(III), As(V) and As(T) are around seven, four and five times more respectively than that of GAC.
- 2. The increase in percentage removal of both As(III) and As(V) with decrease in As_0 value is less for higher value of As_0 (3000–500 ppb) than that for lower value of As_0 (<500 ppb).
- Under the similar experimental conditions, ~99% of Fe can be removed by GAC-Fe whereas Mn removal is only around 41%.
- 4. Effect of adsorbent particle size on the removal of arsenic and iron is insignificant but for Mn it is significant, which indicates the dominating role of physical adsorption for Mn removal over the electrostatic attraction (chemical adsorption).
- 5. Using GAC-Fe (8 g/l) the arsenic concentration in the treated water can be reduced below 10 ppb and 50 ppb from arsenic solutions containing maximum As₀ value of 200 ppb and 520 ppb, respectively. Hence, GAC-Fe may be used to treat the arsenic contaminated ground water in India and Bangladesh as the treated water satisfies the national standard of these countries and the contamination level in ground

water also varies from 50 to 300 ppb. At the same time it may be used for treating arsenic contaminated ground water of developed countries where MCL value of arsenic in drinking water is 10 ppb.

Acknowledgements

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References

- Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), USEPA, 2003.
- [2] P. Mondal, C.B. Majumder, B. Mohanty, Laboratory based approaches for arsenic remediation from contaminated water: recent developments, J. Hazard. Mater. B137 (2006) 464–479.
- [3] WHO 1993, Guidelines for drinking water quality, World Health Organization, Geneva, P-41.
- [4] NHMRC 1996, Australian Drinking Water Guidelines, National Health and Medical Council, Agriculture and Resource Management Council of Australia and New Zealand, Commonwealth of Australia, PF S93.
- [5] European Commission Directive, 98/83/EC, Related with drinking water quality intended for human consumption, Brussels, Belgium, 1998.
- [6] EPA Office of Ground water and drinking water, Implementation guidance for the arsenic rule, EPA report-816-D-02-005, Cincinnati, USA, 1998.
- [7] R. Goel, S.K. Kapoor, K. Misra, R.K. Sarma, Removal of arsenic from water by different adsorbents, Ind. J. Chem. Technol. 11 (2004) 518–525.
- [8] C.P. Huang, L.M. Vane, Enhancing As⁵⁺ removal by a Fe²⁺ treated activated carbon, J. WPCF 61 (1989) 1596–1603.
- [9] Z. Gu, J. Fang, B. Deng, Preparation and evaluation of GAC-based ironcontaining adsorbents for arsenic removal, Environ. Sci. Technol. 39 (2005) 3833–3843.
- [10] L. Roland, J. Vaughan, E.R. Brian, Modeling As(V) removal by a iron oxide impregnated activated carbon using the surface complexation approach, Water Res. 39 (2005) 1005–1014.
- [11] P. Mondal, C.B. Majumder, B. Mohanty, A laboratory study for the treatment of arsenic, iron and manganese bearing ground water using Fe³⁺ impregnated activated carbon: Effects of shaking time, pH and temperature, J. Hazard. Mater. 144 (2007) 420–426.
- [12] L. Zeng, A method for preparing silica-containing iron(III) oxide adsorbents for arsenic removal, Water Res. 37 (2003) 4351–4358.
- [13] M. Pakuła, S. Biniak, A. Swiatkowski, Chemical and electrochemical studies of interactions between iron(iii) ions and an activated carbon surface, Langmuir 14 (1998) 3082–3089.
- [14] S. Goldberg, C.T. Johnston, Mechanisms of arsenic adsorption on amorphous oxides evaluated using macroscopic measurements, vibrational spectroscopy and surface complexation modeling, J. Colloid Interf. Sci. 234 (2001) 204–216.
- [15] M. Vithanage, R. Chandrajith, A. Bandara, R. Weerasooriya, Mechanistic modeling of arsenic retention on natural red earth in simulated environmental systems, J. Colloid Interf. Sci. 294 (2006) 265–272.
- [16] N. Yalcın, V. Sevinc, Studies of the surface area and porosity of activated carbons prepared from rice husks, Carbon 38 (2000) 1943–1945.
- [17] I.A. Katsoyiannis, A.I. Zouboulis, Application of Biological processes for the removal of arsenic from ground water, Water Res. 38 (2004) 17–26.
- [18] M. Edwards, S. Patel, L. McNeill, H. Chen, M. Frey, A.D. Eaton, R.C. Antweiler, H.E. Taylor, Considerations in As analysis and speciation, J. AWWA 90 (1998) 103–113.
- [19] S.L. McGeehan, D.V. Naylor, B. Shafii, Statistical evaluation of arsenic adsorption data using linear plateau regression analysis, Soil Sci. Soc. Am. J. 56 (1992) 1130–1133.

- [20] P. Mondal, C.B. Majumder, Treatment of resorcinol and phenol bearing wastewater by simultaneous adsorption biodegradation (SAB): optimization of process parameters, Int. J. Chem. React. Eng. 5 (2007), S-1-15.
- [21] J. Pattnaik, K. Mondal, S. Mathew, S.B. Lalvani, A parametric evaluation of As(V) and As(III) by carbon based adsorbents, Carbon 38 (2000) 589–596.
- [22] Y. Fujino, T. Tabuchi, H. Yasuda, M. Yamachi, GS Yuasa, Technical Report, 2005, http://www.gs-yuasa.com/jp/technic/vol2/pdf/002_1_032.pdf.
- [23] G. Lihua, C. Longwa, L. Guangming, Y. Jianyi, Studies on the preparation of Fe₂O₃ aerosols through sol-gel process and supercritical drying technique, 2001, http://www.chemistrymag.org/cji/2001/037028pe.htm.
- [24] C. Jing, G.P. Korfiatis, X. Meng, Immobilization mechanisms of arsenate in iron hydroxide sludge stabilized with cement, Environ. Sci. Technol. 37 (2003) 5050–5056.
- [25] J.A. Rodriguez, E. Barrado, M. Vega, F. Prieto, J.L.F.C. Lima, Construction and evaluation of As(V) selective electrodes based on iron oxyhydrox-

ide embedded in silica gel membrane, Anal. Chim. Acta 539 (2005) 229-236.

- [26] S. Musi, G.P. Santana, G. Mit, V.K. Garg, ⁵⁷Fe Mossbauer, FTIR and TEM observations of oxide phases precipitated from concentrated Fe(NO₃)₂ solutions, Cro. Chem. Acta 72 (1999) 87–102.
- [27] H. Visser, C.F. Dube, W.H. Armstrong, K. Sauer, V.K. Yachandra, FTIR spectra and normal mode analysis of a tetra nuclear manganese adamantance-like complex in two electrochemically prepared oxidation states: relevance to the oxygen evolving complex of photo system II, J. Am. Chem. Soc. 124 (2002) 11008–11017.
- [28] C.F. Fuller, J.A. Davis, G.A. Waychunas, Surface chemistry of ferrihydrite, Part 2. Kinetics of arsenate adsorption and co precipitation, Gecochim. Cosmochim. Acta 57 (1993) 2271–2282.
- [29] K.P. Raven, A. Jain, R.H. Loeppert, Arsenite and arsenate adsorption on ferrihydrite: kinetics, equilibrium and desorption envelopes, Environ. Sci. Technol. 32 (1998) 344–349.