The effect of Cl^-, PO_4^{3-}, and SiO_3^{2-} on the adsorption of As(V) and As(III) on bauxite in water

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Abstract

The adsorption performance of bauxite for the removal of As(V) and As(III) from contaminated water was investigated. The effect of initial pH, contact time, and the presence of silicate, phosphate and chloride, at concentrations typically found in the ground drinking water in India/Bangladesh, were investigated. As(V) is adsorbed at a rapid initial rate (>96% adsorption within 1 minute) followed by a slow process, reaching a steady state within 6 hours. In comparison the adsorption of As(III) is slow, only 40% is adsorbed within the first minute that gradually rises to 85% in 6 hours. The presence of chloride has insignificant effect on both As(III) and As(V) adsorption. Silicate and phosphate both significantly affect adsorption of both the arsenic ions. Phosphate affects adsorption more strongly than silicate and their effect on As(III) is higher than As(V). When silicate and phosphate are present together, the adsorption of As(V) is almost the same as for the individual ions. However, the As(III) adsorption is significantly affected. The adsorption is 61% as compared to 73% for silicate and 71% for phosphate when individually present. While chloride alone has insignificant effect on the adsorption of the arsenic ions, it has some influence when present together with silicate or phosphate. Chloride + Phosphate combination increases As(V) adsorption by 3% and As(III) by 8%. Chloride + Silicate combination increases As(V) adsorption by 9% but decreases As(III) by almost the same percentage. As compared to the influence of silicate + phosphate, the combination of the three ions together, lowers the As(V) adsorption by 4% and increases As(III) by 4%.

Keywords: Arsenic removal; Bauxite; Coexisting ions; Adsorption

1. Introduction

Arsenic contamination in water and soil is a worldwide problem (Hingston *et al.*, 2001; Ghosh *et al.*, 2011; Abid *et al.*, 2013). In aqueous media, arsenic exists as arsenate (As(V)) in the form of H_3AsO_4 (pK_{a1} = 2.20, pK_{a2} = 6.97 and pK_{a3} = 11.53), and arsenite (As(III)) in the form of H_3AsO_3 (pK_{a1} = 9.22, pK_{a2} = 12.13 and pK_{a3} = 13.40) (Cornelis, 2005). One of the most common methods for arsenic removal from water is chemical precipitation. However, a fine or amorphous precipitate is difficult to filter. Removal of arsenic by adsorption on a suitable adsorbent is another commonly used method. This method offers several advantages (Mohan and Pittman, 2007). The solid adsorbents can be packed into a column or form a filtration bed to allow contaminated water to pass through. This avoids an extra filtration step and allows reuse of the adsorbent after regeneration (Vagliasindi and Benjamin, 1997; Yuan and Luo, 2001; Yuan *et al.*, 2002). A number of synthetic solid adsorbent materials have been reported, which include pre-formed iron oxides, sand coated with iron oxides, or a polymeric material (polystyrene) (Subramanian *et al.*, 1996; Vagliasindi *et al.*, 1996; Vagliasindi and Benjamin, 1997; Yuan *et al.*, 2002). The use of polymer adsorbents is expensive because of the cost of their manufacture. Natural iron ores containing high iron oxide mineral content have been shown to be promising adsorbents for arsenic (Zhang *et al.*, 2004). Bauxite is another potentially attractive solid adsorbent for removal of arsenic from contaminated water. It is an abundantly available material. The main active components of bauxite ore are iron and aluminium oxide minerals. There are few documented studies on its use as an arsenic adsorptive medium (Baral *et al.*,

2007; Bhakat *et al.*, 2007; Mohapatra *et al.*, 2007; Alshaebi *et al.*, 2009; Wendling and Douglas, 2009). Red mud, the waste product of the 'Bayer' bauxite refining process, has also been shown to be an effective solid adsorbent for removal of arsenic from contaminated water (White *et al.*, 2003).

   In studying adsorbents, account must be taken of the effect of ions which commonly co-exist with arsenic in water and may interfere with the ability of adsorbents to remove arsenic. Silicate and phosphate which commonly occur in groundwater in particular have been shown to reduce the adsorption of arsenic on ferric hydroxide adsorbents (Meng *et al.*, 2000; Jiang, 2001; Singh *et al.*, 2005). The ability of phosphate to compete with arsenate for goethite surface sites is well documented (Parfitt, 1978; Persson *et al.*, 1996). Phosphate, like arsenate, is adsorbed as an inner-sphere complex via a ligand-exchange mechanism. Likewise, numerous investigations have shown that soluble silica also shows high affinity adsorption on surfaces of aluminium and ferric oxides (Hingston and Raupach, 1967; Sigg and Stumm, 1981; Davis *et al.*, 2002). Species which result from Al(III) hydrolysis are known to form complexes with Fe(III) and soluble Si(IV) (Taylor, 1988). A yellow-brown Fe-Al sol is reported to be formed by reaction of a ferrihydrite with the Al hydrolysis species at pH 4–4.2. This solution has been shown to be stable over long periods (Taylor, 1988). Since the functional components for arsenic adsorption on bauxite are iron and aluminium oxides/hydroxides, it is likely that silicate and phosphate would reduce the efficiency of arsenic removal by bauxite. Chloride, which is also generally present at reasonably high concentrations has been shown to reduce significantly the effectiveness of arsenic removal with ion exchangers (Korngolf *et al.*, 2001) but has not been investigated for bauxite.

   This paper reports an investigation of bauxite as an adsorbent for removing arsenic from ground drinking water. The effect of co - presence of silicate, phosphate and chloride ions with As(III) and (V) at concentrations simulating those found in typical ground drinking water of India/Bangladesh was examined. These typical background concentrations in ground water are reported to be. (As(V)=As(III)=0.5 mg/L, silicate=50 mg/L, phosphate=5 mg/L, chloride=180 mg/L (Khoe *et al.*, 1999).

2. Experimental

2.1 Materials: Bauxite was sourced from Alcoa Australia's Worsley bauxite mine, Western Australia. It was first ground to a fine powder using a ring mill grinder and then dry-sieved using mesh screens to produce particles in the range of $65 - 107$ µm diameters. Disodium hydrogen arsenate, arsenic trioxide, sodium dihydrogen phosphate, sodium chloride, sodium hydroxide, and hydrochloric acid were of analytical grade, while sodium metasilicate pentahydrate was laboratory grade. All the reagents were used as received.

2.2 Experimental procedure: For each experiment, stock solutions of As(V) were prepared by dissolving $Na_2HAsO_4.7H_2O$ in Milli-Q water. Stock solutions of As(III) were prepared by dissolving As_2O_3 in Milli-Q water. All the arsenic solutions, particularly As(III), were prepared immediately prior to being used. This was to avoid any oxidation of As(III) which is known to be a slow process (Sumner, 1999). 250 mL conical flasks were used as reactors for the adsorption experiments. For each experiment, unless otherwise stated, 2 g of bauxite (20 g/L) was charged into 100 mL of a solution containing 0.5 mg/L As(V). The same procedure and concentrations were used for experiments involving As(III) as well. Solution pH was manually adjusted to desired values with dilute HCl or NaOH before placing the flasks on a shaking table. HCl was selected to adjust the solution pH in all experiments even though it would introduce Cl⁻ into the solutions. The intention was to avoid introduction of another anion to the solutions by using some other acid. The amount of HCl used to adjust the pH was insignificant and as it will be shown later, chloride had negligible effect on adsorption of the arsenic species investigated in this study. The reaction flasks were capped tightly and immersed in a temperature-controlled water bath (25ºC) and then shaken for the required time at a rate of 120±10 cycle/min with a mechanical shaker (Julabo-SW-20C). Samples were taken at regular intervals and centrifuged at 2500 rpm for 30 minutes, followed by filtration through 0.45 µm membrane filters before sending for analyses of total As, Cl⁻, Si, and P by ICP (VISTA Simultaneous ICP–AES spectrometer, Varian). Standard hydride generation (HG) followed by ICP detection method was used for speciation of As(III) and As(V) in the solution sample (Woodward, 2001).

3. Results and discussion

3.1 Effect of contact time: Figure 1 shows the effect of contact time on arsenic adsorption. For As(V) the amount of residual arsenic in water initially decreased rapidly. About 96% of the initial 0.5 mg/L As(V) was removed within 1 minute (Figure 1), after that As(V) concentration decreased very slowly to below detectable limits (0.01 mg/L) in approximately 180 minutes. Thus, the adsorption kinetics for As(V) was very fast. In comparison, the kinetics for As(III) adsorption was initially fast but then dropped gradually. About 40% of the initial 0.5 mg/L As(III) was removed within 1 minute then gradually increased to 85% in approximately 360 minutes. Thus, bauxite is much less effective in removing As(III) than As(V).
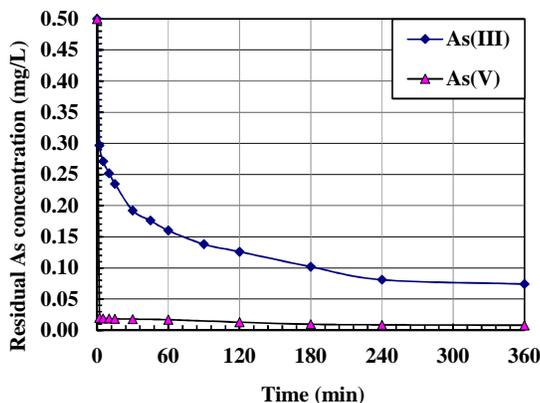
Figure 1. Effect of contact time on arsenic adsorption (temp.=25ºC, initial 0.5 mg/L As(V) or As(III), adsorbent dosage =20g/L).

3.2 Effect of pH: Figure 2 shows the effect of solution pH on As(III) and As(V) adsorption on bauxite. The As(V) removal goes through a maximum in the pH range 5−6. This is not unexpected because at low pH, the iron and aluminium oxides and hydroxides, which are the active components in bauxite for adsorption, undergo partial dissolution, thereby decreasing adsorption sites. At high pH, decrease in As(V) adsorption occurs because OH^- ions compete for adsorption sites on the surface of bauxite. Furthermore the bauxite tends to dissolve via formation of soluble aluminium species at pH higher than 9 (Takeno, 2005). The As(III) adsorption reaches maximum at pH 7.5−8. In comparison to As(V), As(III) adsorption on bauxite was lower and varied with pH in a different manner (Figure 2).
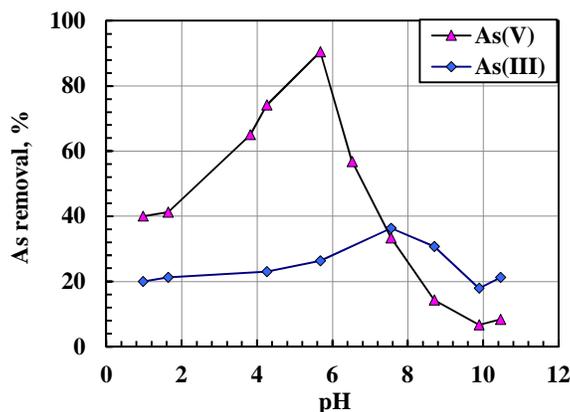

Figure 2. Effect of pH on the adsorption of As(III) and As(V) by bauxite with initial 0.5 mg/L As(V) or As(III).

It is likely that As(V), being a charged species in the range pH 2−9, and hence is easily adsorbed. On the other hand, As(III) is present as neutral arsenous acid in the pH range 2−9 (Takeno, 2005). Hence it is only weakly adsorbed. At pH higher than 9, the adsorption is limited by the tendency of bauxite to dissolve forming soluble iron and aluminium species.

3.3. Effect of chloride: The results of co-presence of chloride in solution on the adsorption of As(V) and As(III) on bauxite are compared in figure 3. As can be seen from the results chloride had negligible effect on As(V) removal. Its residual concentration in solution remains almost zero at all the investigated concentrations of chloride. On the other hand the residual As(III) concentration increased marginally from 70 µg/L to 77 µg/L when chloride was present up to around 50 mg/L. Beyond this the change was negligible. All the added chloride remained in the solution suggesting that adsorption of chloride on bauxite was negligible. It is therefore concluded that chloride has no noticeable effect on the adsorption of As(V), but small effect on As(III). This result is in accordance with literature (Cheng *et al.*, 2015).
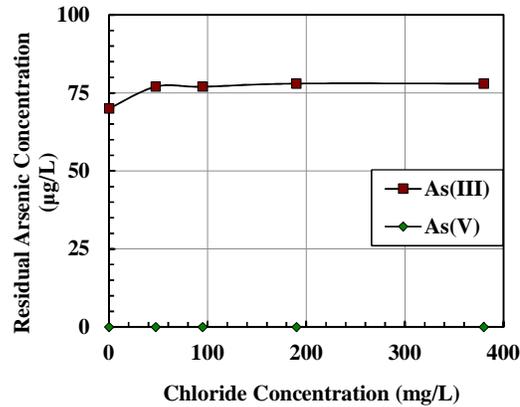
Figure 3. . Effect of chloride on the adsorption of As(III) and As(V) by bauxite (temp.=25°C, initial As(V) 0.5 mg/L, pH=6, initial As(III) 0.5 mg/L, pH=7, adsorbent dosage =20g/L).

3.4 Effect of silicate: The effect of added silicate up to 100 mg/L was investigated in view of its naturally occurring concentration in ground drinking water of India/Bangladesh (Khoe *et al.*, 1999). The results are shown in figure 4 which show that silicate affects adsorption of both As(V) and As(III) quite significantly. As the concentration of silicate is increased, the residual As(V) concentration increases rapidly from zero to 33µg/L up to 10mg/L of added silicate, then it increases gradually to 50µg/L at 100mg/L of added silicate. This means that in the presence of silicate the adsorption of As(V) drops by almost 7% when added silicate concentration in solution is 10mg/L or above. In contrast, the presence of silicate affects the adsorption of As(III) much more strongly. The residual concentration of As(III) increases rapidly from 70 µg/L to 118 µg/L when 10mg/L of silicate is added to the solution. Then it increases gradually to 142µg/L at 100mg/L of silicate. This means that the adsorption of As(III) drops from 86% to 72% (14% drop) at concentrations of added silicate >10mg/L. These results are in general agreement with the findings of others that the presence of silicate in water reduces the effectiveness of arsenic removal by adsorption on other adsorbents such as ferrihydrite (Meng et al., 2000; Singh et al., 2005), iron ores (Zhang et al., 2004) and magnetic iron oxide (Cheng et al., 2015).
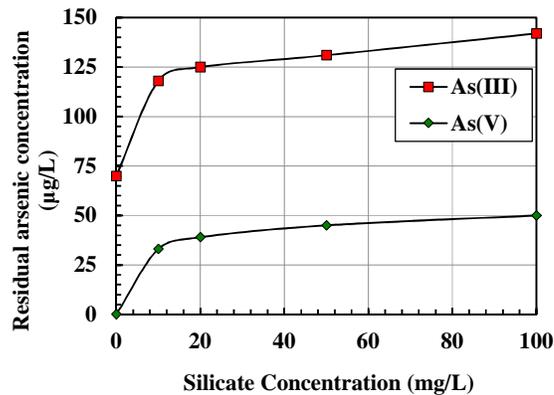

Figure 4. Effect of silicate on the adsorption of As(III) and As(V) by bauxite (temp.=25°C, initial As(V) 0.5 mg/L, pH=6, initial As(III) 0.5 mg/L, pH=7, adsorbent dosage =20g/L).

3.5 Effect of phosphate: The effect of presence of phosphate on the arsenic adsorption on bauxite was investigated under the same conditions as for silicate. As phosphate is added to the solution, the residual concentration of As(V) in solutions increases moderately. As can be seen from the results in figure 5, the addition of up to 5mg/L of phosphate results in the residual As(V) concentration change from almost zero to 15µg/L, meaning that only 3% of the adsorbed As(V) is released into the solution, that is 97% of the As(V) remains adsorbed. At added phosphate concentrations > 5mg/L the residual As(V) concentration gradually increases to 65µg/L. This indicates that with 10mg/L of added phosphate, 87% of the available As(V) remains adsorbed. This is in contrast to 93% which remained adsorbed in case of silicate under similar conditions. Thus phosphate affects the arsenic adsorption more than silicate.
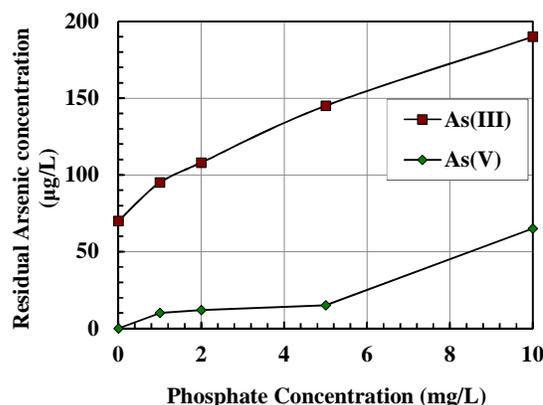
Figure 5. Effect of phosphate on the adsorption of As(III) and As(V) by bauxite (temp.=25ºC, initial As(V) 0.5 mg/L, pH=6, initial As(III) 0.5 mg/L, pH=7, adsorbent dosage =20g/L).

Figure 5 also shows the effect of added phosphate on As(III) adsorption. With the increase of phosphate from zero to 10mg/L the residual As(III) concentration increases linearly from 70μg/L to 190μg/L. This indicates that the adsorption of As(III) decreases progressively from 86% to 62%, representing 24% drop when 10 mg/L of phosphate are added to the solution. This is in contrast to the drop of 14% observed for silicate noted in section 3.4. This result is consistent with that reported by Cheng et al (Cheng *et al.*, 2015). Phosphate is known to be strongly adsorbed on bauxite (Kamiyango *et al.*, 2013). Thus, phosphate competes with arsenic species for the adsorption sites on bauxite.

3.6. Effect of chloride, silicate, and phosphate combination: Since chloride, silicate, and phosphate are present together *in* groundwater, it is important to investigate the combined effect of these ions on arsenic adsorption at concentrations simulating those actually found in ground water (Khoe *et al.*, 1999). The results of the investigation are shown in Table 1.

Table 1 Comparison of arsenic adsorption from water containing various combinations of the interfering ions. (As(V)=As(III)=0.5 mg/L, silicate=50 mg/L, phosphate=5 mg/L, chloride=180 mg/L, bauxite= 20 g/L, pH =7)

Combinations	%As(V) ads.	%As(III) ads.
No interfering ions	100	85
Chloride	100	84
Silicate	91	73
Phosphate	97	71
Silicate + Chloride	100	64
Phosphate + Chloride	100	79
Silicate + Phosphate	93	61
Silicate + Phosphate + Chloride	89	65

93% of As(V) is adsorbed when silicate and phosphate are present together. This is almost an average of the individual effects of these ions, 97% and 91% respectively for phosphate and silicate when present alone. The adsorption of As(III) is 61% when silicate and phosphate are present together as compared to 73% for silicate and 71% for phosphate when they are present alone. Thus, about 10% less As(III) is adsorbed when both the ions are present together. When chloride is present with either phosphate or silicate or silicate + phosphate combination, it displays a noticeable effect on both the arsenic ions adsorption. Chloride + phosphate combination increases As(V) adsorption by 3% and As(III) by 8%. Chloride + silicate combination increases As(V) adsorption by 9% and decreasing As(III) by the As(V) adsorption increased by 9%, and that of As(III) decreased by almost the same percentage. When chloride is combined with phosphate and silicate present together the As(V) adsorption decreases from 93% to 89%, a reduction of 4%. Whereas the A(III) adsorption increases from 61% to 65% ie an increase of 4%.

4. Conclusions

The removal of As(III) and As(V) from water by adsorption on bauxite under various conditions is investigated. The effect of co-presence of chloride, silicate and phosphate on adsorption of the two arsenic ions has also been examined. It is found that an adsorption maximum for As(V) occurs in the pH range 5-6 and that for As(III) in the range 7.5-8. The adsorption kinetics of both

the arsenic ions is initially fast. About 96% of As(V) is removed within the first minute and then slowly increases to 100% in 180 minutes of contact time. Only 40% of As(III) is removed within the first minute which gradually changes to 85% in 360 minutes. Thus bauxite is a much better adsorbent for As(V) than for As(III). The presence of chloride alone has almost no effect on the adsorption of both the arsenic ions. The presence of each of phosphate and silicate ion individually decreases adsorption of both the arsenic ions. The effect is dependent on the concentration of the added ions. In the co-presence of 10mg/L silicate, As(V) adsorption drops by almost 7% and As(III) by 14%. In the presence of chloride, silicate and phosphate at concentrations which simulate their concentrations in the ground drinking water in India/Bangladesh, it is found that while chloride alone has insignificant effect on the adsorption of the arsenic ions, it does have some influence when present together with silicate and phosphate. Chloride + Phosphate combination increases As(V) adsorption by 3% and As(III) by 8%. Chloride + Silicate combination increases As(V) adsorption by 9% but decreases As(III) by almost the same percentage. When the three ions are present together, As(V) adsorption decreases by 11% and As(III) by 20%.

Acknowledgement

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