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Removing arsenic from water by coprecipitation with iron: Effect of arsenic and iron concentrations and adsorbent incorporation

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Abstract

Arsenic (As) contamination of drinking water is a major cause of As toxicity in many parts of the world. A study was conducted to evaluate As removal from water containing 100-700 $\mu\text{g/L}$ of As and As to Fe concentration ratios of 1:5 – 1:1000 using the coprecipitation process with and without As/Fe adsorption onto granular activated carbon (GAC). Fe concentration required to reduce As concentrations in order to achieve the WHO standard level of 10 $\mu\text{g/L}$ increased exponentially with the increase in initial As concentration. When small amounts of GAC were added to the As/Fe solutions the Fe required to remove these As concentrations reduced drastically. This decline was due to the GAC adsorption of Fe and As, enhancing the removal of these metals through coprecipitation. Predictive regression

equations were developed relating the GAC dose requirement to the initial As and Fe concentrations. Zeta potential data revealed that As was adsorbed on the GAC by outer-sphere complexation whereas Fe was adsorbed by inner-sphere complexation reversing the negative charge on GAC to positive values. X-ray diffraction of the GAC samples in the presence of Fe had an additional peak characteristic of ferrihydrite (Fe oxide) compared to that of the GAC sample without Fe. The study showed that incorporating an adsorbent into the coprecipitation process has the advantage of removing As from waters at all concentrations of Fe and As compared to coprecipitation alone which does not remove As to the required levels if Fe concentration is low.

Keywords: adsorption, arsenic, coprecipitation, granular activated carbon, water treatment

1. Introduction

Arsenic (As) contamination of drinking water is a serious water quality problem in many parts of the world, especially Vietnam, Bangladesh, India, Taiwan, Cambodia, and Chile [1-3]. Continued drinking of groundwater contaminated with As for many years can cause skin ailments such as hypopigmentation (white spots on skin), hyperpigmentation (dark spots on skin), keratosis (break-up of the skin on hands and feet), and melanoma [4-6]. Groundwater As concentration in many parts of the above-mentioned countries have reached levels of more than 10-100 times the WHO's maximum permissible concentration of 10 $\mu\text{g/L}$ [6-8].

Two common methods of removing As from drinking water are adsorption and coprecipitation with Fe [1-3,9-15]. The latter method is a natural process where the concentration of Fe found in groundwaters generally increases with As concentration [16]. This increase is mostly due to As in anoxic groundwater originating from the anoxic

dissolution of As-rich iron oxyhydroxides that occur as dispersed phases in the aquatic rocks [8,17]. The efficiency in removing As from groundwater by coprecipitation depends on the Fe/As ratio – the higher the Fe/As ratio the better the removal efficiency [8]. Very high ratios were found to be essential for efficient As removal. However, very little quantitative information is available on the Fe/As ratio required for reducing the As concentration to the WHO standard level for waters containing diverse As and Fe concentrations [13,14].

At low Fe concentration, As cannot be easily removed by coprecipitation because Fe does not form a precipitate that can adsorb the As [13,14]. If there is insufficient Fe in the water for As coprecipitation, an adsorbent could be incorporated into the coprecipitation process so that Fe adsorption and/or surface precipitation is induced, which will promote As removal. Such a technique has not been tested previously. The objectives of this study are as follows. Firstly, to determine quantitative relationships of As/Fe ratio requirements necessary for the removal of As from waters which contain diverse amounts of As concentrations. Secondly, to determine the amount of GAC adsorbent required for removing As from waters with different As and Fe concentrations using the adsorption/coprecipitation hybrid process. Previous studies considered adsorption and coprecipitation separately and there have been numerous studies conducted on adsorptive removal of As using several adsorbents, especially the iron-based adsorbents [11]. If the proposed hybrid process of incorporating GAC into coprecipitation is found to be successful, it would have the advantage of removing As from waters regardless of the concentrations of Fe and As. An additional advantage is the removal capacity of As could be much higher than the adsorptive capacity of adsorbents used alone without the inclusion of coprecipitation process. GAC is used as the adsorbent because it is a popular and efficient adsorbent for removing As [3] and many other co-existing contaminants from water [18-20]. Furthermore, Fe oxyhydroxide doped activated carbon has already been shown to have high adsorptive capacity for As [9,11] and As coprecipitation with Fe in the

presence of GAC would behave like the Fe-doped activated carbon or even better because of the surface precipitation of high concentrations of Fe and As on GAC which leads to higher As removal capacity.

2. Materials and methods

2.1. Materials

GAC with a particle size of 0.3–2.4 mm was obtained from James Cummins P/L, Australia. A narrow particle size range of 300-600 μm was separated by sieving the original material and the sieved material used for the study. The experiments tested a range of As (V) concentrations (100 - 700 $\mu\text{g/L}$) and As/Fe ratios (1:5 - 1: 1000) with and without the addition of various doses of GAC to cover the differing levels of As contaminated groundwaters in most of the As contaminated countries. Analar-grade $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ obtained from Sigma-Aldrich, USA was used in the experiments.

2.2. As removal by coprecipitation with Fe

The feed solution was prepared at As concentrations of 100, 200, 300, 500, and 700 $\mu\text{g/L}$ using $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$. Coprecipitation experiments were conducted with different As to Fe ratios (1:5 to 1: 1000). The Fe solution was prepared using $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$. To different volumes of As solution in 250 mL flasks, varying amounts of FeSO_4 solution containing 100 mg Fe/L were added and the total volume was made to 100 mL. The flasks were agitated in a shaker for 24 h at 120 rpm at room temperature ($23 \pm 1^\circ\text{C}$). The contents in the flasks were filtered using a 1.2 μm filter and the filtrates analysed for As and Fe using an Inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7900). The amounts of As and Fe removed in the precipitates were subsequently determined.

2.3. As removal by coprecipitation and adsorption on GAC in the presence of Fe

Different amounts of GAC were added (0.05 – 1.0 g/L) to solutions containing As and Fe concentrations (As/Fe ratio 1:5 - 1:1000) as prepared in the coprecipitation experiment (section 2.2) in 250 mL flasks, and the contents agitated in a shaker for 24 h at 120 rpm at room temperature ($23 \pm 1^\circ\text{C}$). The contents were filtered and analysed as described in the previous section. The amounts of As and Fe removed by GAC were then determined.

2.4. Zeta potential

Zeta potential is an important parameter used to understand the mechanism of adsorption because it is the electrical potential close to a particle's surface where adsorption of ions from the solution phase occurs and is positively related to the surface charge. The zeta potential values were measured on 100 mL suspensions containing 1.0 g/L of GAC alone; GAC with 200 $\mu\text{g/L}$ As; GAC with 10 mg/L of Fe; and GAC with 200 $\mu\text{g/L}$ As and 10 mg/L Fe. These were conducted all in the presence of 10^{-3} M of NaCl at the pH range of 5.0 - 8.0 using a Zetasizer nano instrument (Nano ZS Zen3600, Malvern, UK). The measurements were recorded after the suspensions at the different pHs were agitated in a shaker for 24 h. The pH was measured after 4 h shaking, readjusted back to the initial pH and the shaking continued. pH after 24 h was also measured.

2.5. Chemical compounds formed during coprecipitation

X-ray diffraction (XRD) analysis was conducted using a PANalytical Empyrean instrument operated at 60-kV with Cu-Ka1 radiation on powdered samples of all precipitates to determine the different minerals formed during the coprecipitation process. The mineral

compositions of GAC + As, GAC + Fe and GAC + Fe + As were compared with that of GAC. The samples used for XRD were prepared by coprecipitation using a solution of 250 mg/L of Fe and 500 $\mu\text{g/L}$ of As in the presence of GAC (1 g/L). Not enough solid precipitate was collected from the suspension formed by coprecipitation of As and Fe in the absence of GAC. For this reason, XRD was not conducted on this sample.

3. Results and discussion

3.1. Coprecipitation

The results of the coprecipitation experiments showed that As concentrations fell when a decrease in the As/Fe ratio occurred. This is due to increased amounts of Fe hydroxide precipitate formed at high Fe concentrations coprecipitated with As. The data for initial As concentrations of 500 $\mu\text{g/L}$ are shown in Fig. 1 as an example and for other As concentrations (100, 200, 300, and 700 $\mu\text{g/L}$) are presented in Fig. S1-S4. The As/Fe ratio required to reduce As concentration to the WHO level decreased (Fe concentration increased) when As concentration increased (Fig. 1.a, Fig. S1.a-S4.a). It was 1:45, 1:70, 1:300, 1:400 and 1:500 for the initial As concentrations of 100, 200, 300, 500, and 700 $\mu\text{g/L}$, respectively. For a fixed initial As concentration, as the As/Fe ratio decreased, the final As concentration decreased but Fe concentration increased. The decrease in As concentration levels is due to its removal during coprecipitation. Although Fe was also removed in proportion to the removal of As, there was excess Fe in the solution and this increased as the As/Fe ratio decreased.

Arsenic removed in coprecipitates can get resuspended on holding the coprecipitate. To avoid this problem the resuspended or unremoved As after coprecipitation can be removed by adsorption columns as post treatment process using appropriate adsorbents (1-3, 10,11). Otter et al. [21] used such a technique by filtering the coprecipitate through MnO_2

coated sand packed in a column in a continuous process and backwashed the column when the precipitate accumulated in the filter to prevent filter clogging. At village level in Bangladesh, As is removed by coprecipitation in bucket treatment units where the coprecipitate formed is allowed to settle down to the bottom of the bucket and the treated water containing As below the toxic level is collected from taps installed few cm above the bottom of the bucket before resuspension occurred [14].

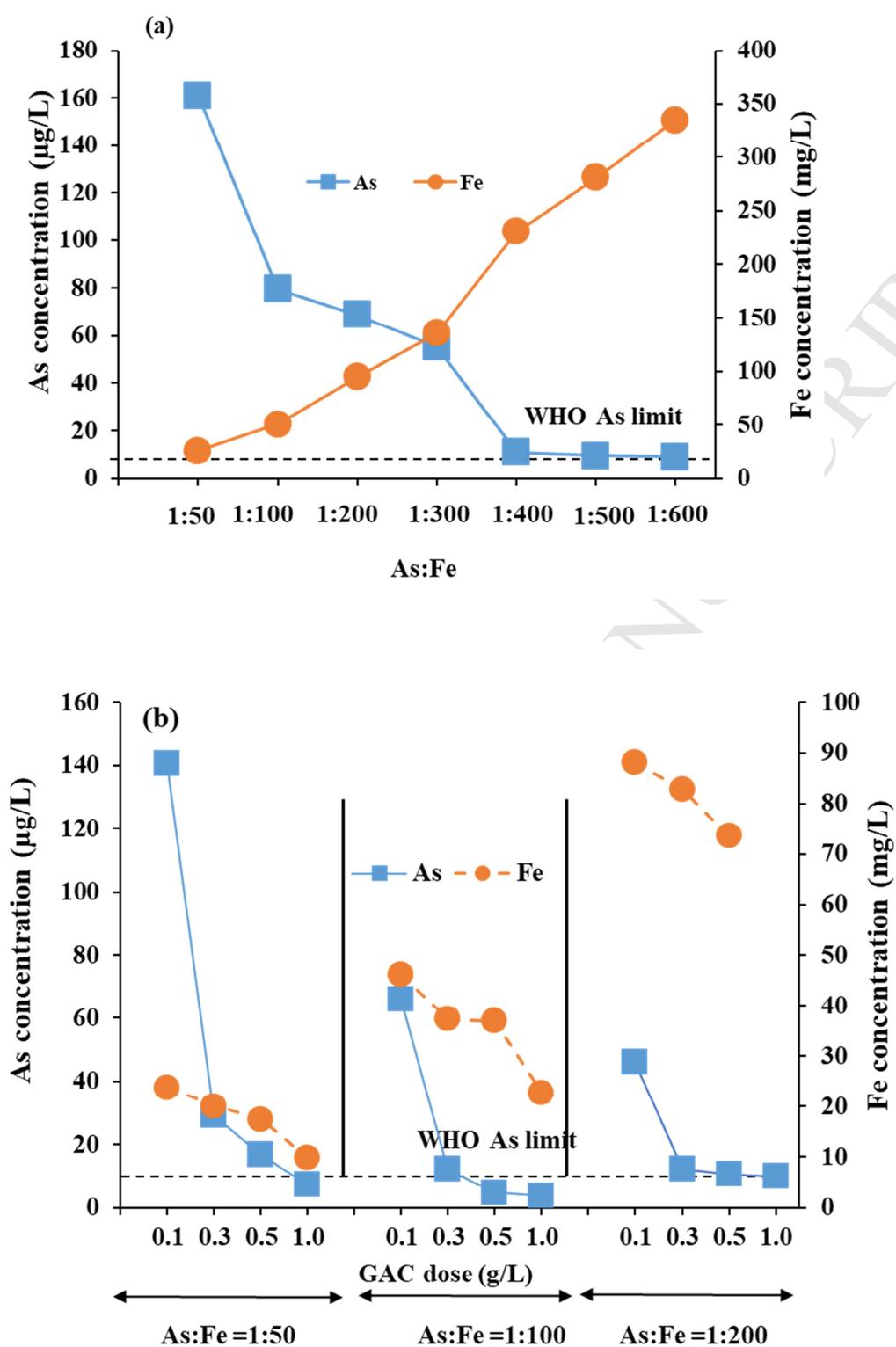


Fig. 1. Effect of As:Fe ratio in water on the efficiency of As removal using As/Fe coprecipitation at an As concentration of 500 µg/L (a) without and (b) with the addition of GAC.

3.2. Adsorption/coprecipitation

The results of the adsorption/coprecipitation experiments demonstrated that when a small amount of GAC was added to the As/Fe solution, the As/Fe ratio required for As concentration reduction drastically increased (i.e. reduction of Fe requirement). For example, without the addition of GAC, when the As concentration was 500 $\mu\text{g/L}$, an As/Fe ratio of 1:400 was required to reduce the As concentration to the WHO level by coprecipitation. However, by adding a GAC dose of 1 g/L, it was possible to reach the WHO's recommended level at an As/Fe ratio of 1:50 (Fig. 1). The required GAC dose increased when the As concentration rose and As/Fe ratio decreased (Fig. 1.b, Fig. S1.b-S4.b). The final concentrations of both As and Fe were lower in the presence of GAC rather than when GAC was absent. This shows that GAC adsorption of Fe and As enhanced the coprecipitation removal capacity of these metals. The results also indicated that the removal of As and Fe by the adsorption/coprecipitation process increased at a larger GAC dose (Fig. 1.b, Fig. S1.b-S4.b).

The As removal capacity by GAC adsorption/coprecipitation depends on the initial As concentration, As:Fe ratio and GAC dose (Table 1). It increased with increase in initial and equilibrium As concentrations and decrease in GAC dose and As:Fe ratio. The maximum adsorption/coprecipitation capacity obtained within the range of parameters used in the study was 6.38 mg/g (for initial As concentration 0.7 mg/L, equilibrium As concentration 0.061 mg/L, As:Fe ratio 1:200, and GAC dose 0.1 g/L) (Table 1). This value is much higher than the maximum adsorption capacity reported for other adsorbents for similar initial or equilibrium As concentrations. For example, Hao et al. [11] reviewed the adsorption capacities of several iron-based adsorbents and reported that the capacities ranged from 0.2 to 2.5 mg/g when the initial As concentrations were ≤ 1 mg/L. Similarly, Gula et al. [10] reported adsorption capacities of 0.02-2.8 mg/g for several activated carbons and iron-loaded

activated carbons for initial As concentrations ≤ 1 mg/L. Gu et al. [9] studied the adsorption of As on four types of Fe impregnated GACs ranging in Fe contents of 11-23 mg/g and reported that their adsorption capacity at equilibrium As concentration of 1 mg/L increased with Fe content from 0.1 to 3.5 mg/g.

Table 1. As adsorption/coprecipitation capacity at different initial As concentrations, As/Fe ratios and GAC doses.

Initial As concentration (mg/L)	As:Fe ratio	GAC dose (g/L)	Equilibrium As concentration (mg/L)	As adsorption/coprecipitation capacity (mg/g)
0.1	1:5	0.05	0.012	2.86
		0.10	0.009	1.46
		0.30	0.008	0.49
		0.50	0.006	0.30
	1:10	0.05	0.011	2.88
		0.10	0.008	1.47
		0.30	0.006	0.50
		0.50	0.003	0.31
0.2	1:5	0.05	0.023	3.86
		0.10	0.014	2.02
		0.30	0.007	0.70
		0.50	0.006	0.42
		1.00	0.003	0.21
	1:10	0.05	0.016	3.99
		0.10	0.010	2.06
		0.30	0.003	0.71
0.3	1:10	0.30	0.014	1.07
		0.50	0.011	0.65
		1.00	0.009	0.33
	1:30	0.30	0.012	1.08
		0.50	0.008	0.66
		1.00	0.005	0.34
	1:50	0.30	0.002	1.11
		0.50	0.001	0.67
1.00		0.00008	0.36	
0.5	1:50	0.10	0.141	4.56
		0.30	0.029	1.89
		0.50	0.017	1.16
		1.00	0.008	0.59
	1:100	0.10	0.066	5.31
		0.30	0.012	1.95
		0.50	0.005	1.18
		1.00	0.004	0.60
0.7	1:200	0.10	0.029	5.68
		0.30	0.008	1.96
		0.50	0.007	1.19
		1.00	0.006	0.61
	1:50	0.10	0.240	4.58
		0.30	0.061	2.13
		0.50	0.031	1.34
		1.00	0.011	0.69
0.7	1:100	0.10	0.126	5.73
		0.30	0.031	2.23
		0.50	0.013	1.37
		1.00	0.008	0.70
	1:200	0.10	0.061	6.38
		0.30	0.016	2.28
		0.50	0.007	1.38
		1.00	0.004	0.71

3.3. Regression analysis to determine mathematical relationships

As the final As concentration (y_1) ($\mu\text{g/L}$) in water after coprecipitation treatment depends on the initial As concentration before the treatment (x_1) ($\mu\text{g/L}$) and initial Fe concentration before the treatment (x_2) ($\mu\text{g/L}$), two types of multiple regression analysis were conducted to determine mathematical relationships between these parameters. The first type had no squared term in the analysis and the resulting regression equation was:

$$y_1 = 257 x_1 - 0.78 x_2 + 0.72 x_1 x_2 - 10.06 \quad (R^2 = 0.61) \text{----- (1)}$$

Because the relationships were curvilinear (Fig. 1.a, Fig. S1.a-S4.a), the multiple regression analysis was repeated by including squared terms for the independent variables to improve the relationship. This analysis resulted in the following equation with a higher R^2 value. Therefore, this equation (equation 2) is considered to be a better predictive tool for managing As in water by the coprecipitation method. It explains 74% of the variation in the data.

$$y_1 = 333 x_1 - 0.61 x_2 - 0.25 x_1 x_2 + 62.78 x_1^2 + 0.0006 x_2^2 - 28.54 \quad (R^2 = 0.74) \text{----- (2)}$$

Berg et al. [12] reported an empirical relationship of the percentage As removal from groundwaters of Vietnam containing 10-382 $\mu\text{g/L}$ of As and 0.1-48 mg/L of Fe by coprecipitation and Fe concentration using the equation $\text{As removal (\%)} = 13.6 \ln (\text{Fe, mg/L}) + 45$ which indicate that very high Fe concentrations are required for reducing As concentration to low levels. They stated that the concentration of Fe in groundwater was the decisive factor for the effective removal of As. Mamtaz and Bache [14] conducted a

regression analysis to determine the Fe concentration required to reduce the As concentration in synthetic waters to the Bangladesh standard limit of 0.05 mg/L. They found a curvilinear relationship of $Fe = 66 As^{1.75}$ (units of As and Fe concentrations is mg/L) but the R^2 value or any other measure of the closeness of data fit was not reported for this relationship. Using a similar analysis, we also found a strong curvilinear relationship between Fe concentration required to reduce As concentration to the WHO limit of 10 $\mu\text{g/L}$ ($Fe = 1337 As^{2.51}$, $R^2 = 0.97$) (unit of As and Fe is mg/L) (Fig. 2).

The relationship between Fe concentration required to reduce As concentration to the standard limit in our study is different from that reported by Mamtaz and Bache [14] because of two main reasons. One reason is that the standard As limit used to derive the relationship in our study is the WHO limit of 0.01 mg/L, whereas in the study of Mamtaz and Bache [14] the standard As limit used was 0.05 mg/L. The other reason is that As[III] species was used in the study of Mamtaz and Bache [14] but As(V) was the As species used in our study. The difference in the relationships could also be due to the dissimilarity in the methodology used in the two studies. However, both relationships reveal that the Fe concentration requirement increased exponentially with initial As concentration, and therefore, it may not be possible to reduce As levels to the WHO recommended limit if the Fe concentration is not sufficiently large enough.

The Fe concentration required to reduce As concentration to the Bangladesh standard limit of 0.05 mg/L can be obtained for the initial As concentrations of 100, 200, and 300 $\mu\text{g/L}$ in our study from Fig. S1.a, Fig. S2.a and Fig.S3.a, respectively. They are 0.2, 6, and 70 mg/L, respectively. The corresponding Fe concentration requirement reported in the study of Mamtaz and Bache [14] were 1, 5, and 9 mg/L, respectively. This analysis reveals that the range of Fe concentration requirement is similar in both studies at low As concentrations but much higher in our study at high As concentrations. In Vietnam, using raw groundwater

containing As (10-382 $\mu\text{g/L}$) and Fe (0.1-48 mg/L), it was reported that Fe/As ratios of ≥ 50 were required to ensure As removal to 0.05 mg/L standard level [12]. However, the absolute Fe concentration required to meet this standard was not provided in the study.

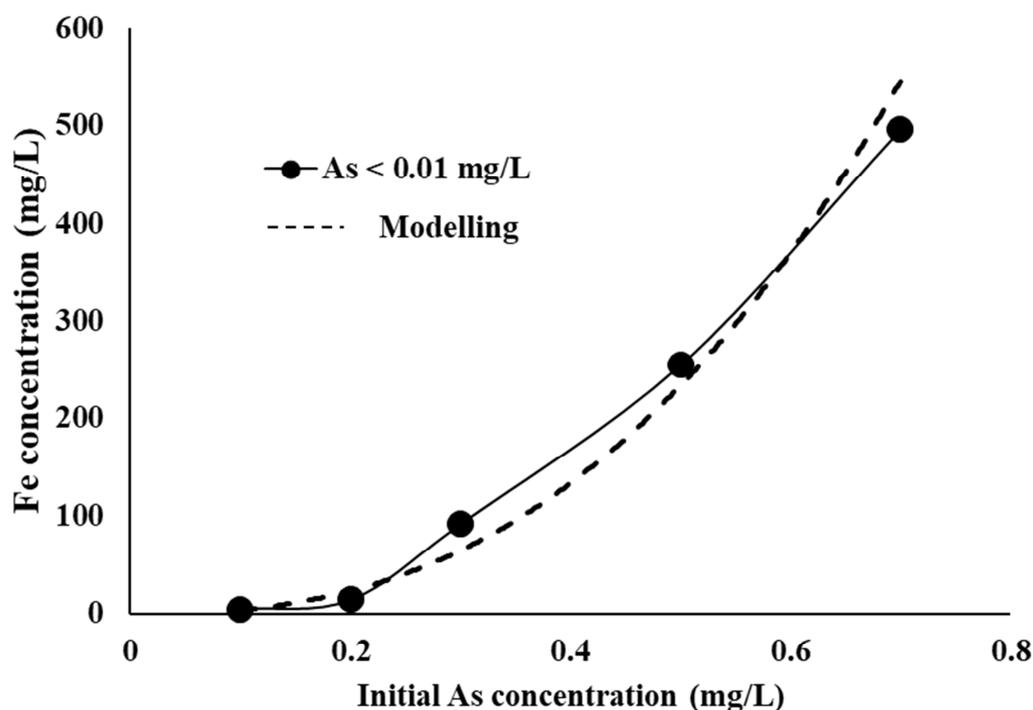


Fig. 2. Fe concentration required to reduce the initial As concentration to 10 $\mu\text{g/L}$.

Adding a small dose of GAC to the coprecipitation process helped to reduce the Fe concentration (or increase As/Fe ratio) requirement to reduce As concentration to the recommended WHO limit (Fig. 1.b, Fig. S1.b-S4.b). Therefore, multiple regression analyses were conducted to determine the GAC dose (y_2) (g/L) required to reduce the As concentration to the WHO level of 10 $\mu\text{g/L}$ for the different initial As and Fe concentrations. The equations obtained were:

$$y_2 = 2.22 x_1 - 0.02 x_2 + 0.0203 x_1 x_2 - 0.21 \quad (R^2 = 0.82) \text{ -----(3)}$$

$$y_2 = 4.73 x_1 - 0.03 x_2 + 0.018 x_1 x_2 - 2.29 x_1^2 - 4.23 * 10^{-5} x_2^2 - 0.63 \quad (R^2 = 0.88) \text{ -----(4)}$$

Of the two regression equations the second one (equation 4) is a better predictive tool for determining the GAC dose in a coprecipitation/adsorption treatment process. This is because of its higher R^2 value and it explains 88% of the variation in data.

3.4. Zeta potential

Zeta potential data show that GAC is negatively charged and this negative charge increased with pH (Fig. 3), indicating that As ion adsorption on the GAC with negatively charged adsorption sites by simple electrostatic attraction (outer-sphere coordination) would be hindered because of electrical repulsion. However, the zeta potential of GAC in the presence of As decreased (negative zeta potential increased) suggesting that As has been adsorbed probably by inner-sphere coordination (chemical adsorption) to the GAC surface. The zeta potential in the presence of Fe with or without As increased and became positive at pHs less than 6.5. This indicates that Fe is also adsorbed by inner-sphere coordination but producing surface positive charges as reported for other heavy metals [19]. These data show that when GAC was added to water containing Fe and As, it can adsorb both Fe and As and provide a conducive surface for Fe and As coprecipitation. The zeta potential of GAC in the presence of As was the lowest, but when Fe was added, it produced the highest zeta potential and became positive at pHs less than 6.5. This was due to the positive charges created on GAC by Fe adsorption. Therefore, the positively charged Fe may have helped the adsorption of As to produce some form of Fe arsenate compound.

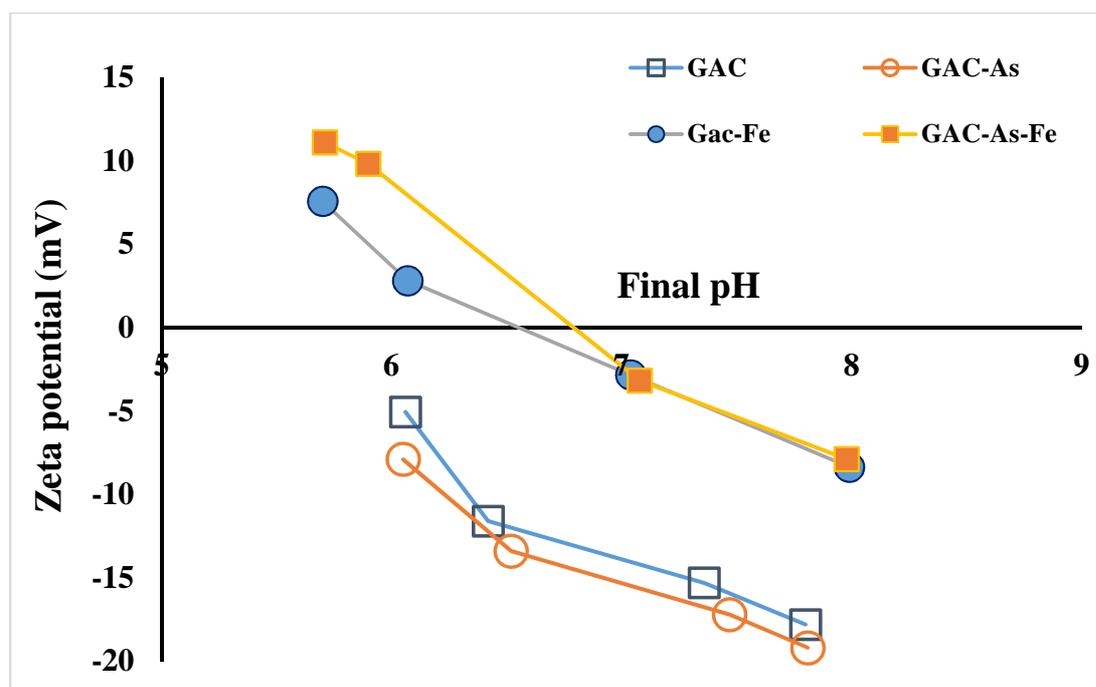


Fig. 3. Zeta potential of GAC (1 g/L) in the presence of 200 $\mu\text{g/L}$ of As and 10 mg/L of Fe as a function of pH.

3.5. pH changes during adsorption/coprecipitation

When GAC was added to the As/Fe solution the pH changed mostly during the first few hours. Therefore, after 4 h of shaking the suspensions, the pHs were noted down and readjusted back to the initial pH, and the adsorption process continued for 24 h. At low initial pHs, the pH increased during the first 4 h (ΔpH positive) (Fig. 4) due to adsorption of protons (H^+) from the acidic conditions (low pH) containing abundant protons, leaving more OH^- ions in solution. The ΔpH decreased at intermediate pHs and became negative at high pHs. This is probably because protons were released from the functional groups of GAC at high pHs. The ΔpH was lower in the presence of Fe (GAC-Fe, GAC-As-Fe) compared to that in the absence of Fe (GAC, GAC-As). This is probably because Fe adsorption on GAC by

inner-sphere complexation as indicated by the zeta potential data in the previous section would have released H^+ into solution resulting in pH reduction.

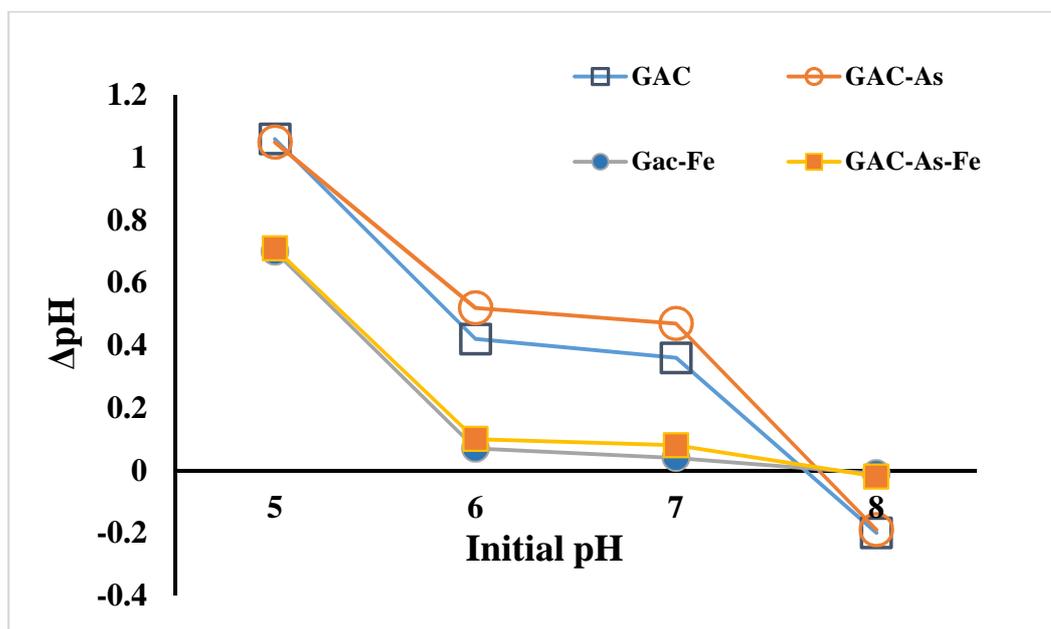


Fig. 4. pH changes during adsorption/coprecipitation ($\Delta\text{pH} = \text{final pH} - \text{initial pH}$) of Fe/As on GAC (1 g/L) in the presence of 200 $\mu\text{g/L}$ of As and 10 mg/L of Fe as a function of initial pH.

3.6. XRD analysis

XRD analysis was conducted to determine the different minerals formed during the coprecipitation/adsorption process. The mineral composition of GAC was compared with that of GAC + As, GAC + Fe and GAC + Fe + As (Fig. 5). The XRD pattern of GAC showed strong crystalline peaks at 2θ values of 21.1, 26.8, 36.7, 44.9 and 50.3 which are characteristics of quartz (SiO_2) mineral as also found for other GACs [22]. These peaks are due to the quartz impurity found in the commercial GAC. These peaks were also detected in the GAC + As, GAC + Fe, and GAC + Fe + As samples. However, the GAC + Fe and GAC + Fe + As samples which contained Fe had an additional peak

characteristic of ferrihydrite (Fe oxide) at 2θ of 35.7 [23]. This indicated that the Fe added to GAC in the As coprecipitation/adsorption process had formed a Fe oxide precipitate, probably ferrihydrite on the GAC surface consistent with the zeta potential data. Others have also reported this peak in Fe impregnated GAC/carbon samples and ascribed this as due to Fe_3O_4 precipitate without naming the mineral formed [22,24]. However, Gallios et al. [25] could not find any XRD peak associated with Fe in samples of AC impregnated with Fe oxide. They explained this as being due to insufficient Fe in AC or the Fe oxide formed was amorphous (non-crystalline) which cannot be measured by XRD. Fig. 5 shows that no new peak formation occurred in the GAC + Fe + As sample compared to the GAC + Fe sample. Indicated here is that As had not formed any crystalline compounds with Fe on the GAC. Alternatively, had there been any Fe/As compound formation, it was amorphous and cannot be detected by XRD.

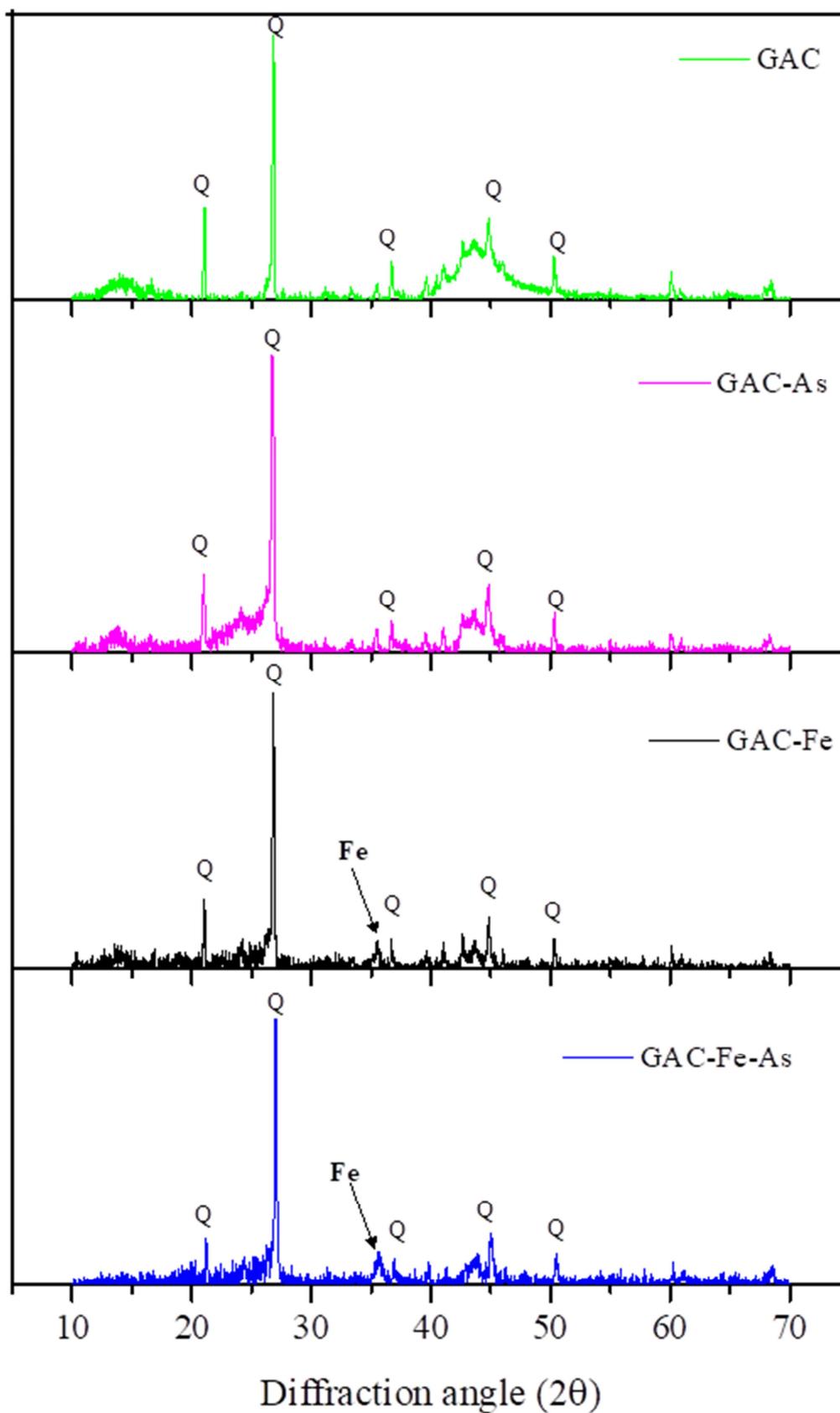


Fig. 5. XRD patterns of GAC, GAC + As, GAC + Fe, and GAC + Fe + As (Q: quartz, Fe: ferrihydrite).

4. Conclusions

Coprecipitation of As with Fe was able to reduce initial As concentration in water to the WHO level of 10 $\mu\text{g/L}$ at all initial As concentrations of 100 – 700 $\mu\text{g/L}$ tested. However, the Fe concentration required to reduce the As concentration increased (As/Fe concentration ratio reduced) against the As concentration in an exponential pattern, so much so, that an As/Fe ratio of 1/500 was required for the initial As concentration of 700 $\mu\text{g/L}$ compared to an As/Fe ratio of 1/40 for the initial As concentration of 100 $\mu\text{g/L}$. Adding a small dose of 1 g/L of GAC adsorbent was able to reduce the As concentration from an initial 700 $\mu\text{g/L}$ to the WHO limit at an As/Fe ratio of 1:50. The higher the dose of GAC the better the efficiency of As concentration reduction and a smaller Fe concentration was required (higher As/Fe ratio). A possible reason for this is the higher adsorption of Fe on GAC, which promotes increased coprecipitation of As with the adsorbed Fe.

Zeta potential of GAC in the presence of Fe was positive compared to the negative zeta potential of GAC alone or with As at pHs less than 6.5. The adsorption of Fe might have produced surface Fe oxy-hydroxide precipitate on GAC. This was supported by the formation of an XRD peak characteristic of ferrihydrite for GAC + Fe samples. The study indicated that the adsorbent GAC when it is incorporated into the coprecipitation process has the advantage of removing As from waters at all concentrations of Fe and As compared to coprecipitation on its own which will not remove As to the required levels if the Fe concentration is low.

We recommend future studies on investigating whether the residual As left in water at low Fe concentrations/or low GAC doses in the coprecipitation/adsorption process can be removed by adding low doses of GAC or Fe-doped GAC after the coprecipitation process.

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SUPPLIMENTARY DATA**Removing arsenic from water by coprecipitation with iron: Effect of arsenic and iron concentrations and adsorbent incorporation**

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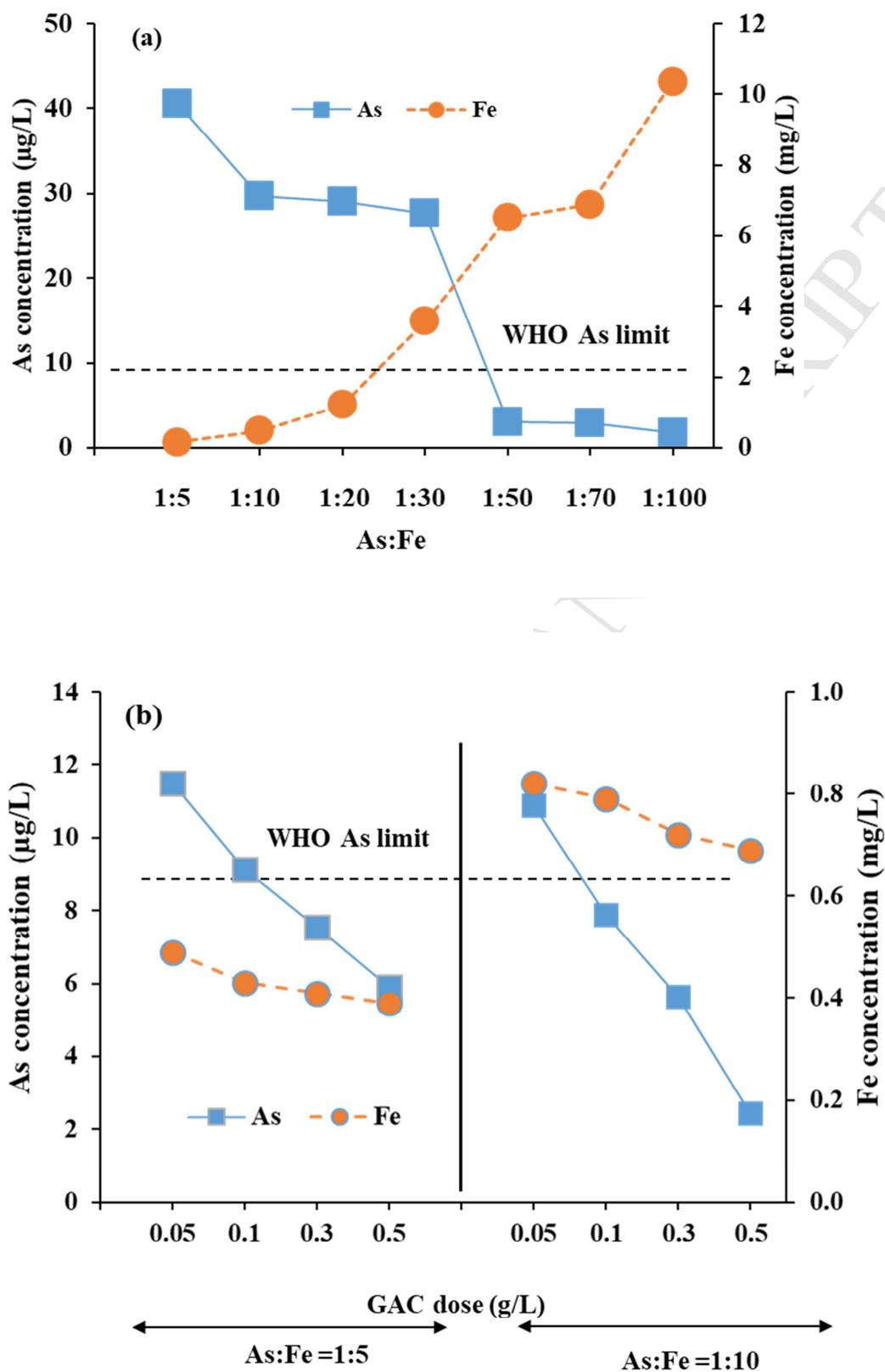


Fig. S1. Effect of As:Fe ratio in water on the efficiency of As removal by As/Fe coprecipitation at an As concentration of 100 µg/L (a) without and (b) with addition of GAC.

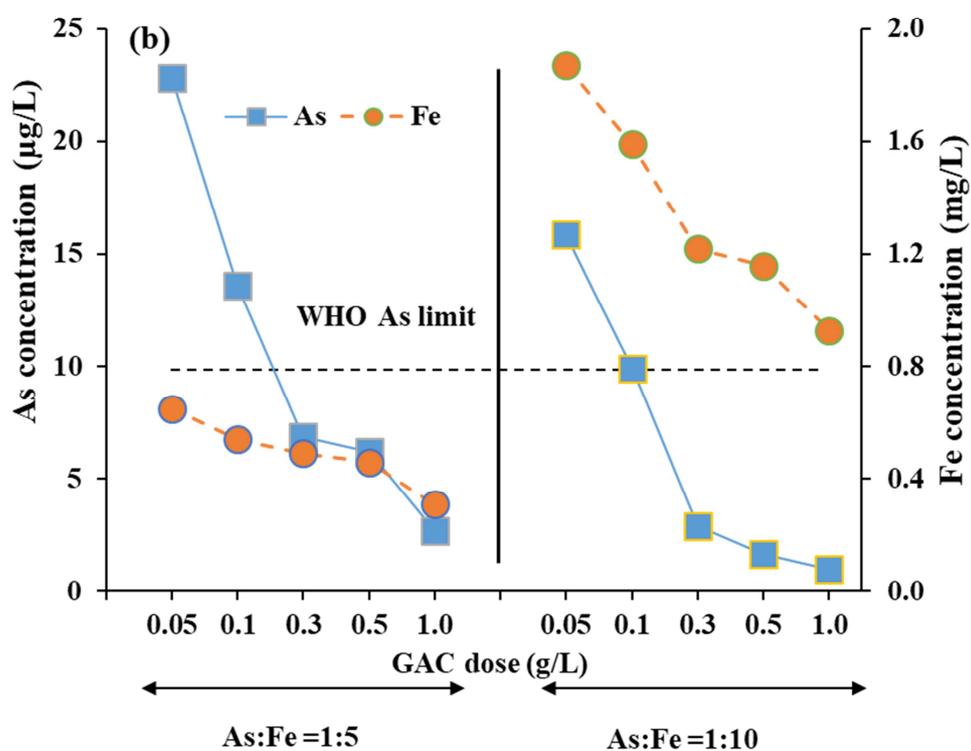
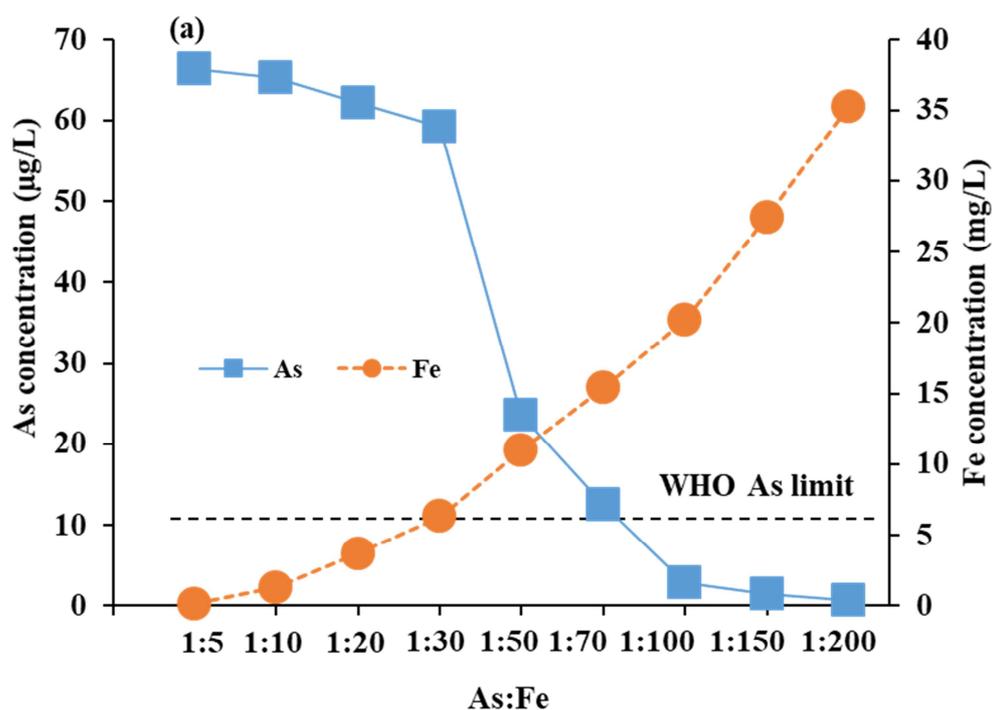


Fig. S2. Effect of As:Fe ratio in water on the efficiency of As removal by As/Fe coprecipitation at an As concentration of 200 µg/L (a) without and (b) with addition of GAC.

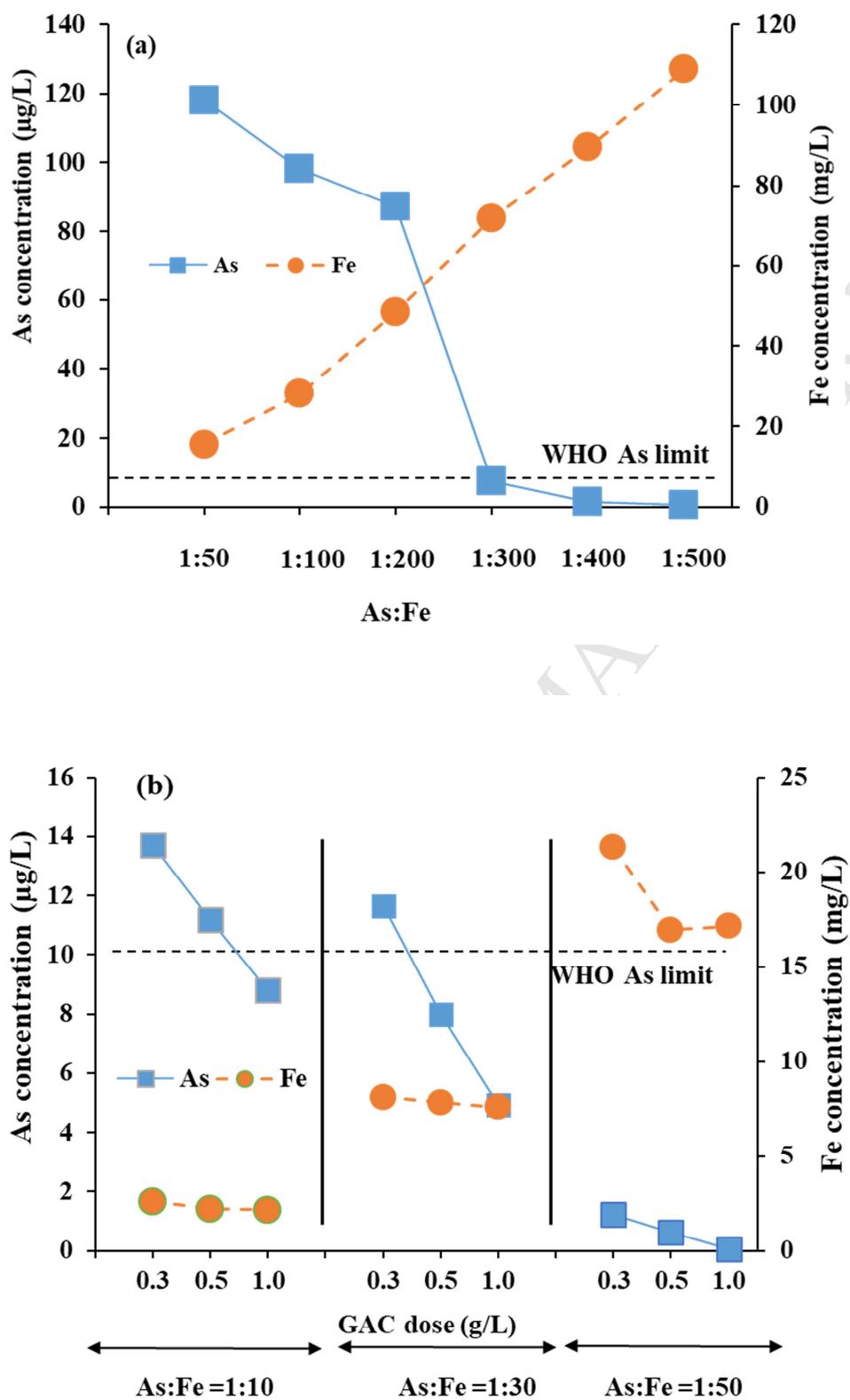


Fig. S3. Effect of As:Fe ratio in water on the efficiency of As removal by As/Fe coprecipitation at an As concentration of 300 µg/L (a) without and (b) with addition of GAC.

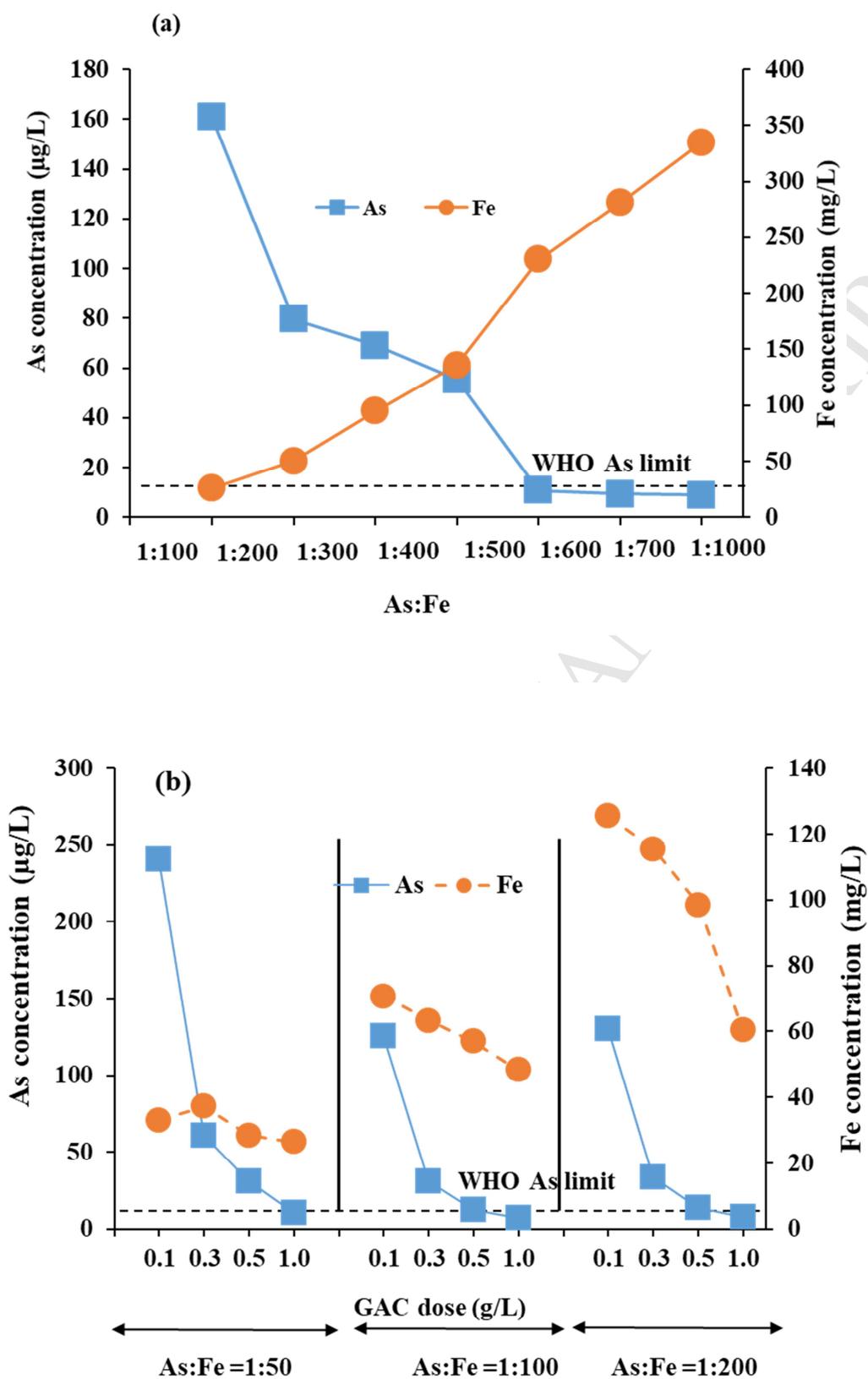


Fig. S4. Effect of As:Fe ratio in water on the efficiency of As removal by As/Fe coprecipitation at an As concentration of 700 µg/L (a) without and (b) with addition of GAC.

Highlights:

- As/Fe coprecipitation effectively removed As from water only at high Fe levels.
- At low Fe levels, Fe/As adsorption on GAC/coprecipitation effectively removed As.
- Fe levels required for coprecipitation increased exponentially against As levels.
- High Fe levels changed GAC surface negative charges to positive values.
- XRD data showed precipitation of Fe oxyhydroxide on GAC surface at high Fe levels.