



Coagulation of arsenic adsorbed ferrihydrite with the use of polyaluminium chloride (PAC) or polyferric sulfate (PFS)

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Received 10 October 2011; Accepted 15 March 2012

ABSTRACT

Ferrihydrite is effective in arsenic removal because of the considerable amount of active hydroxyl groups. Solid–liquid separation of the arsenic adsorbed ferrihydrite from aqueous solution is important for the arsenic treatment technology. Coagulation is a promising process for ferrihydrite separation. Effects of different coagulants on arsenic adsorbed ferrihydrite settlement were investigated. Surface charge significantly decreased after arsenic was adsorbed on ferrihydrite. Turbidity, iron and arsenic removal efficiencies were used to characterize the ferrihydrite settling process. Turbidity removal efficiency with polyferric sulfate (PFS) added was 98.2% when pH was 5.0. Meanwhile, the turbidity removal rate with polyaluminium chloride (PAC) added was 96.3–97.3% when the pH of colloidal suspensions was 7.0–9.0. Arsenic or iron removal rate after 30 min settling was improved from about 40 to 80% with coagulants added. The mean size of flocs after coagulation process was 61.8 μm after PFS was added, or 71.6 μm after PAC was added when the pH was 6.5. The floc structure of ferrihydrite became more compact and stable with PAC or PFS added.

Keywords: Coagulation; Ferrihydrite; Arsenic adsorbed

1. Introduction

Arsenic is considered to be a hazardous metal in terms of its detrimental impact on human beings and ecological systems. It occurs naturally in several minerals, which can release soluble arsenic into natural waters [1]. Arsenic can also be found in the waste streams from industries such as smelting, petroleum, and metallurgical processes [2]. Arsenic is also released into the environment by the dispersion of arsenic-containing fertilizers, pesticides, and wood

preservatives. The World Health Organization sets the maximum permissible concentration for arsenic in drinking water at 10 μg/l. However, long term exposure through drinking water to even low concentrations of arsenic (≤ 50 μg/l) can cause carcinogenic diseases of skin, lungs, blood and kidneys [3].

In order to meet the rising demand of standard drinking water, there is growing interest for the development of novel materials or technologies for arsenic removal from drinking water or industrial effluents before it may cause significant contamination. Although many different methods

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such as co-precipitation by aluminum or iron hydroxides, adsorbing colloid flotation, ion-exchange, and membrane filtration have been used for arsenic removal [4–6], the process based on co-precipitation technology is promising, due to high arsenic removal efficiency and low cost.

Co-precipitation of arsenic with iron has been widely studied as a means of removing arsenic from drinking waters and wastewaters [7,8]. There have been extensive studies on the co-precipitation of arsenate with iron(III) regarding the process, the solubility and stability of the arsenic-bearing co-precipitates, the speciation of arsenic and the parameters influencing these properties. Co-precipitation process with iron added generated iron hydroxide precipitates, which were disposed as sludge waste.

Traditionally, iron hydroxide precipitates encountered in the co-precipitation process have been described as amorphous ferric hydroxide, colloidal ferric hydroxide, $\text{Fe}(\text{OH})_3$, etc. However, the precipitates do not have the composition $\text{Fe}(\text{OH})_3$, and they are not amorphous although their X-ray diffracting properties are poor. It was recognized that many of these precipitates are actually ferrihydrite [9].

Ferrihydrite is the mineral name identifying poorly ordered hydrous ferric oxide precipitates, which have stoichiometry near $\text{Fe}_2\text{O}_3 \cdot 2\text{H}_2\text{O}$ [10]. Lots of researches have reported that ferrihydrite has a tremendous adsorption capacity for aqueous arsenic [11,12]. 0.7 mol As(V)/mole Fe was achieved when iron was co-precipitated in the presence of As(V) in the pH range 7.5–9.0. Meanwhile, adsorption of As(V) by ferrihydrite was 0.25 mol As/mole Fe by solid slurry adsorption.

Considering the high arsenic adsorption capacity of ferrihydrite, during the co-precipitation process for arsenic treatment, recycling ferrihydrite could decrease the required dosages of iron and thus reduce the amount of sludge. The traditional and modified arsenic removal processes are shown in Fig. 1A and B. Settlement of recycling flocs during coagulation process was studied before [13]. However, the effects of arsenic on ferrihydrite coagulation in sector 2# of Fig. 1B were not reported.

Inorganic polymer flocculants, such as polyaluminum chloride (PAC) and polyferric sulfate (PFS) are widely investigated in synthetic colloidal systems and in water treatment. The aim of the present study is to investigate the coagulation process of arsenic adsorbed ferrihydrite in the presence of PFS or PAC. A study of their flocculation kinetics and floc structures in aggregation can help in the better understanding of flocculation mechanisms. Flocculation kinetics of ferrihydrite with arsenic adsorbed induced by these

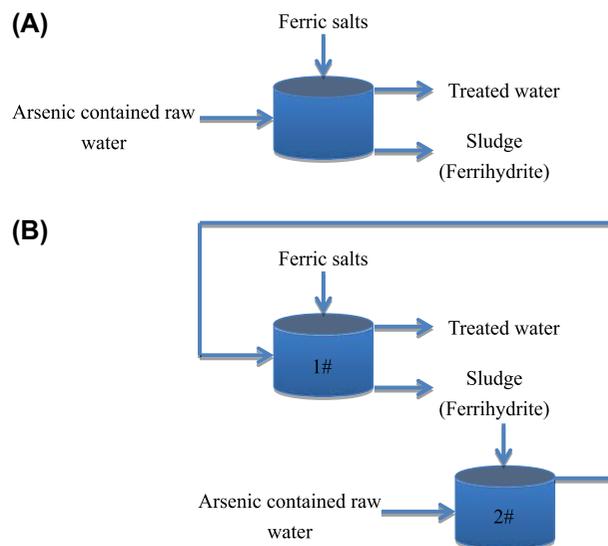


Fig. 1. Arsenic removal process with (B) or without (A) ferrihydrite recycling.

two polyelectrolytes was compared. Their floc structures are studied and the flocculation mechanisms are also discussed.

2. Material and methods

2.1. Materials and reagents

Considering that CO_3^{2-} was usually rich in arsenic containing water, ferrihydrite was synthesized in the laboratory described as follows. To 500 ml of a solution containing 1 M of Na_2CO_3 , 1 M FeCl_3 was added at a fixed rate of addition of approximately 100 ml/min, during vigorous stirring with a magnetic stirrer. The pH of the suspension was then adjusted to 7.5 by the dropwise addition of 1 M FeCl_3 . The sediment was washed three times with deionized water and separated from solution by centrifugation at 5,000 rpm for 10 min. The centrifugation sediment was re-suspended in deionized water, diluted to 200 ml final volume, and stored at room temperature. The ferrihydrite concentration in the final suspension was approximately 0.1 g/ml. The freeze-dried sample was analyzed by powder X-ray diffraction using Cu K α ($\lambda = 0.15418$ nm) radiation at 40 kV and 30 mA from a Shimadzu XRD-6000 diffractometer (Japan), and the 2-line ferrihydrite was confirmed. The results of XRD analysis of 2-line ferrihydrite were widely reported in previous studies. Meanwhile, the results of this work showed little discrepancy with the dates reported [14–16]. Therefore, the results of XRD analysis of ferrihydrite were not shown in the manuscript. It was reported that ferrihydrite transformed partially into goethite upon

prolonged storage (>4 weeks at room temperature) [17]. Therefore, each batch of ferrihydrite was used in the batch studies within 7 days of its synthesis.

2.2. Coagulation/flocculation experiments

Coagulation/flocculation experiments were carried out by using the Jar Test method. The Numeric Flocculator JT-M6 (A-choice instrument Co., Ltd, USA) was equipped with stainless-steel paddles (7.5×2.5 cm) and allowed to work simultaneously with six 1-L tall-form cylindrical beakers. The coagulation experiments were conducted at room temperature ($23\text{--}26^\circ\text{C}$). An amount of 0.1 g ferrihydrite was first added in 1 L deionized water. $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ salt (Merck 6284) was dissolved in water for 50 mg/l As (V) stock solution. These stock solutions were used to prepare experimental solutions of specified concentrations with deionized water. A certain concentration of arsenic was then added in the cylindrical beaker. The mixture was stirred under 300 rpm for 1 h. In this way, simulated recycled ferrihydrite with arsenic adsorbed was prepared.

The influence of coagulant dose on arsenic adsorbed ferrihydrite removal was studied by using PFS or PAC as coagulant at concentrations between 0 and 100 mg/l. (PAC: Al_2O_3 10%, PFS: Fe 11%). pH was fixed at different values (with NaOH 0.5 M), from 5.0 ± 0.2 to 9.0 ± 0.2 , to study its influence on arsenic adsorbed ferrihydrite removal under these specific coagulant conditions.

Appropriate contact times consist of 3 min of rapid mixing (300 rpm) for the coagulation step, 15 min of slow mixing (70 rpm) for the flocculation step, and 30 min without mixing for the settling step. The flocs were allowed to settle for 30 min, and the initial (after rapid mixing) and residual (after settling) turbidity was measured using a HACH 2100N IS Turbidimeter (USA). Meanwhile, iron and arsenic concentrations were measured during settling step. Samples were digested by Aqua regia (3:1 HCl:HNO₃) in a microwave oven (MARS, CEM, USA). The ferrihydrite settling rate in the coagulation process was characterized as turbidity, arsenic, and iron removal rates.

2.3. Analytical methods

The total arsenic was analyzed with an Atomic Fluorescence Spectrometer (AF-610A, Rayleigh Analytical Instrument Corp. China). Iron concentration was measured by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES, Prodigy, Leeman LABS, USA). Zeta potential measurements were realized on colloidal suspensions with a Zeta potential

analyzer (Zetasizer 2000, Malvern Co., UK). The adsorption concentration of arsenic on ferrihydrite was 20 mg/g, the ion strength was not artificially controlled. A certain amount of coagulant was added at the beginning of the coagulation process. Samples were collected by a plastic syringe during the coagulation process and tested immediately. The time for one measurement was set to 1 min to record the zeta potential distribution and one sample was measured twice. The pH was adjusted to the prescribed value during the measurement. Size distributions and mass structural information of flocs were determined using a Malvern Mastersizer 2000 (Malvern Co., UK), which ascertains the size by analysis of forward scattered light. The suspension was measured by continuous recycling of water flowing through the sample cell of the instrument. A peristaltic pump with 5 mm internal diameter at a flow rate of 2.0 l/h was used to draw the suspension. The pump was located downstream of the instrument to prevent disturbing the flocs prior to measurement. The test solution and pH was artificially controlled in accordance with the coagulation test.

Mass fractal dimension was measured using Small-Angle Light Scattering as described in the literatures before [18,19]. This method involves the measurement of light intensity I as a function of the scatter vector Q . The vector is defined as the difference between the incident and scattered wave vectors of the radiation beam in the medium, which is given by

$$Q = \frac{4\pi n}{\lambda} \sin(\theta/2) \quad (1)$$

where n , θ , and λ are the refractive index of the medium, the scattered angle, and the wavelength of radiation in vacuum, respectively. The mass fractal dimension D_f can be determined from the negative slope of a plot of log scatter intensity as a function of log scatter vector:

$$I \propto Q^{-D_f} \quad (2)$$

3. Results and discussion

3.1. Effect of arsenic on the coagulation process of ferrihydrite

The turbidity removal rates measured during settling process with the aforementioned system are shown in Fig. 2. Ferrihydrite removal with PAC or PFS added in settlement process could be at rapid stage (0–10 min) followed by a slow stage (10–20 min). An

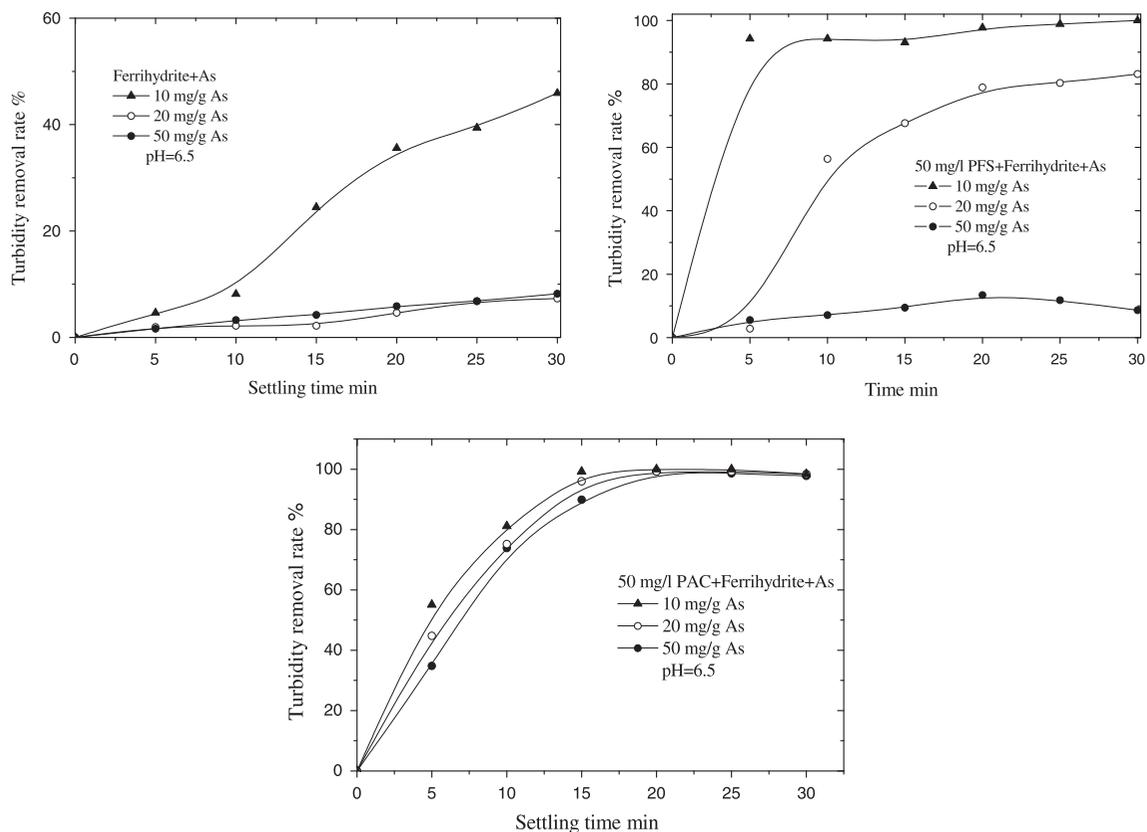


Fig. 2. Removal of turbidity by coagulation process as a function of time.

increase in the settling time above 20 min gives little further effect on settling of ferrihydrite.

The effect of arsenic concentration on turbidity removal was conducted when the pH of colloidal suspensions was adjusted to 6.5. Arsenic has significant effect on turbidity removal without coagulants added or with PFS added, more arsenic inhibited ferrihydrite settling. Turbidity was effectively removed with PAC aided techniques when pH was 6.5. When 10 mg/g arsenic was adsorbed on ferrihydrite, turbidity could be effectively removed after 30 min settling with PFS or PAC aided techniques. Arsenic and iron concentration were also measured to characterize the arsenic adsorbed ferrihydrite removal during settling. The initial iron concentration with or without PFS added in Fig. 3 was 55.13 ± 5.12 mg/l or 49.91 ± 5.12 mg/l. The results of arsenic and iron removal rates during ferrihydrite settling shows similar trends with turbidity removal. When the pH was adjusted to 6.5, ferrihydrite with 20 mg/g arsenic adsorbed was more effectively removed with PAC aided techniques compared to PFS or no coagulants added.

The corresponding stable species of pentavalent arsenic was H_2AsO_4^- or HAsO_4^{2-} when the pH was 6.5. The reaction of arsenic and ferrihydrite could be

as complex, Jain et.al. reported the possible reactions of arsenic adsorption of ferrihydrite [20], more amount of arsenic adsorbed could lead to more negative charges onto the surface of ferrihydrite. The results were in accordance with Fig. 4 that the surface charge decreased significantly after arsenic was adsorbed on the surface of ferrihydrite. Surface charge of colloidal particles was one of the most important factors which controlled the coagulation process.

3.2. Effects of coagulation pH, zeta potential and coagulants dosages on the coagulation of ferrihydrite

The removal of colloidal ferrihydrite with arsenic adsorbed by PFS or PAC was related to the solution pH. Under the same dosage of coagulant, a lower removal rate was observed at higher solution pH when PFS was added (Fig. 5). However, increased pH favors ferrihydrite removal when PAC was used as the coagulant.

It was reported that PFS is more positively charged at low pH [21]. Zeta potential of colloidal ferrihydrite decreased with more arsenic adsorbed. The coagulation results with PFS added were in accordance with Fig. 4 that more amount of arsenic

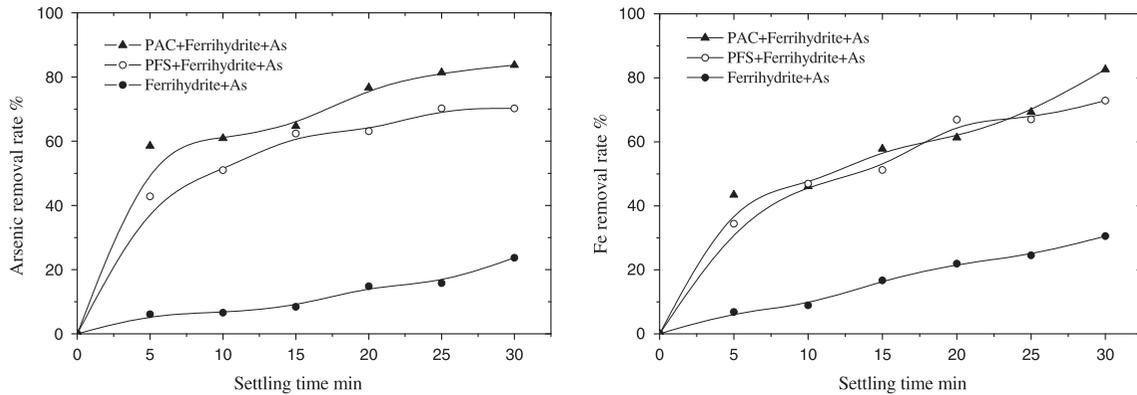


Fig. 3. Removal of arsenic or iron by the coagulation process as a function of time. Samples were digested with Aqua regia; the adsorption concentration of arsenic on ferrihydrite was 20 mg/g; and the dosages of PAC and PFS were 50 mg/l.

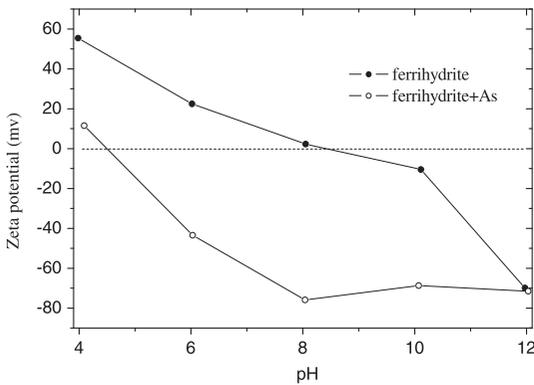


Fig. 4. Zeta potentials of ferrihydrite as a function of pH. The adsorption concentration of arsenic on ferrihydrite was 20 mg/g.

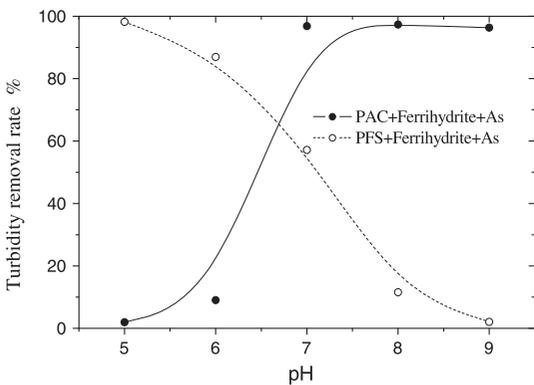


Fig. 5. Removal of turbidity by coagulation as a function of pH; the concentration of arsenic adsorbed on ferrihydrite was 20 mg/g; the dosages of PAC and PFS were 50 mg/l.

adsorbed decreased the removal efficiency of ferrihydrite. As pH increased, more $\text{Fe}(\text{OH})_3$ flocs or $\text{Fe}(\text{OH})_4^-$ ions formed, which reduced the chance for

colloidal ferrihydrite to associate with PFS. The pH increase did not favor ferrihydrite removal as the zeta potential of colloidal suspension decreased (Fig. 5).

Since the negative charge on ferrihydrite with arsenic adsorbed preferably conjugates the PFS, the complexes formed will precipitate out of the solution through charge neutralization. It can be concluded that the removal rate of ferrihydrite is higher at acidic solution. However, it was shown in Fig. 6 that at low pH there is also not enough positive charge on PFS to neutralize the negative charge on arsenic adsorbed ferrihydrite. Sorption and bridging could also be important in coagulation of ferrihydrite with PFS. It was reported that when pH was higher than 6.0, adsorption reactions at the surface of ferric hydroxides and co-precipitation with ferric hydroxides predominate, removal of colloidal suspensions by sorption

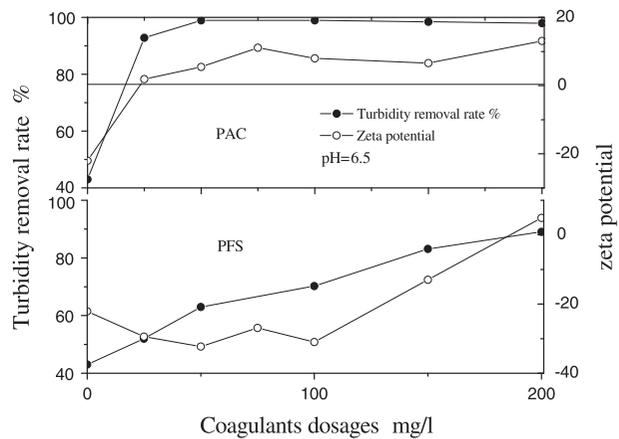


Fig. 6. The effects of coagulants dosages on turbidity removal and the zeta potential in the coagulation process. The concentration of arsenic adsorbed on ferrihydrite was 20 mg/g.

was limited and hydroxide precipitates were poorly sorbing compounds [22]. When the pH of colloidal suspension was controlled at 6.5, the arsenic adsorbed ferrihydrite acquired such a high level of negative charge that the PFS is unable to neutralize it completely. In this condition, increasing the dosages of PFS could provide more positive charges thus improving the arsenic adsorbed ferrihydrite settling rate (Fig. 6).

The results in Fig. 6 showed that PAC was more effective in neutralizing the negative charge on the surface of colloidal ferrihydrite than PFS. The pH value ranging from 7.0 to 9.0 was of advantage for arsenic adsorbed ferrihydrite removal when 50 mg/l PAC was added (Fig. 5). The colloidal particles could not effectively aggregate in $\text{pH} < 6$ with PAC added. The reasons may be as follows. Firstly, the negative charge on colloidal ferrihydrite rapidly changed to positive charge stable particles after PAC was added. Secondly, it was reported that effective coagulation of PAC was in the narrow pH range for acidic circumstances. H^+ inhibited the coagulation process of the multi-hydroxyl polymer of PAC. When the pH of colloidal suspensions was higher than 7, the positive charge of PAC could effectively neutralize the negative charge on the surface of arsenic adsorbed ferrihydrite. In this way, the removal efficiency of the colloidal particles was high in the pH range of 7.0–9.0.

The turbidity removal rate was higher than 90% after 25 mg/l PAC was added when the pH was 6.5. The removal efficiency of colloidal ferrihydrite slightly increased after more PAC was added and showed little changes when the dosage of PAC was higher than 50 mg/l. The efficiency of coagulation efficiency of

PAC was higher than PFS when the pH of colloidal suspension was 6.5. The effects of PAC or PFS concentration on the zeta potential of colloidal particles were shown in Fig. 6 and the pH was artificially controlled at 6.5. After 25 mg/l PAC was added, the zeta potential of the colloidal arsenic adsorbed ferrihydrite increases significantly from -22 to 1.9 . However, the zeta potential was negative and showed little changes when the dosages of PFS were lower than 100 mg/l.

3.3. Particle size and structure of flocs

The floc size distribution of colloidal ferrihydrite with arsenic adsorbed was shown in Fig. 7. The mean particle size $D(4,3)$ and fractal dimension of the particles were shown in Table 1. As shown in Fig. 7 and Table 1, the particle size of intermediate flocs increased after PAC or PFS added. The results were in accordance with Fig. 2 that coagulants were of advantage in ferrihydrite removal.

It was reported that the rate of aggregation of particles depends on several factors, such as primary particle concentration, collision efficiency, and mode of particle transport [23]. With the aforementioned system (i.e. the same concentration of primary colloidal ferrihydrite particles and mixing conditions), the kinetics of aggregation should thus depend mainly on the collision efficiency. The high removal efficiency of colloidal ferrihydrite by PAC at pH 6.5 can be partially explained by the surface charge neutralization, hence, increased particle collision efficiency.

Information about the ferrihydrite floc structures can be obtained by analyzing the data from static light scattering. D_f values in different coagulation times

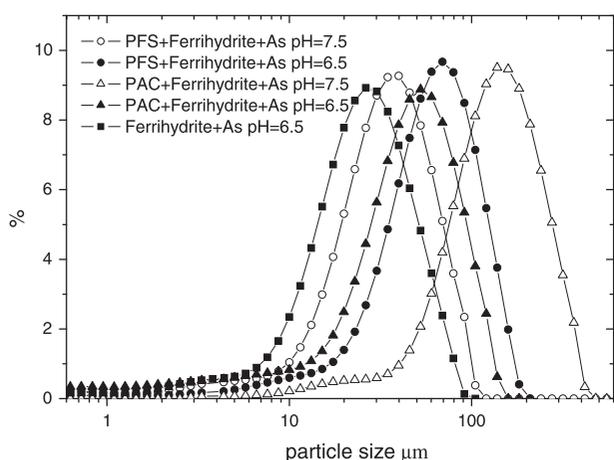


Fig. 7. Size distributions of ferrihydrite particles in the coagulation process (50 mg/l PAC or PFS added, 20 mg/g As adsorbed).

Table 1

Particle size and fractal dimensions in different times of the coagulation process

Flocculants	Time (min)	Particle size (μm)	Fractal dimensions D_f
PFS pH = 6.5	3	18.5	1.9
	10	58.6	2.1
	18	61.8	2.2
PFS pH = 7.5	3	17.6	1.7
	10	28.6	1.9
	18	35.6	2.1
PAC pH = 6.5	3	14.2	1.4
	10	40.9	1.6
	18	45.0	1.9
PAC pH = 7.5	3	71.6	1.8
	10	110.9	2.0
	18	138.9	2.2
Colloidal ferrihydrite	18	13.5	1.2

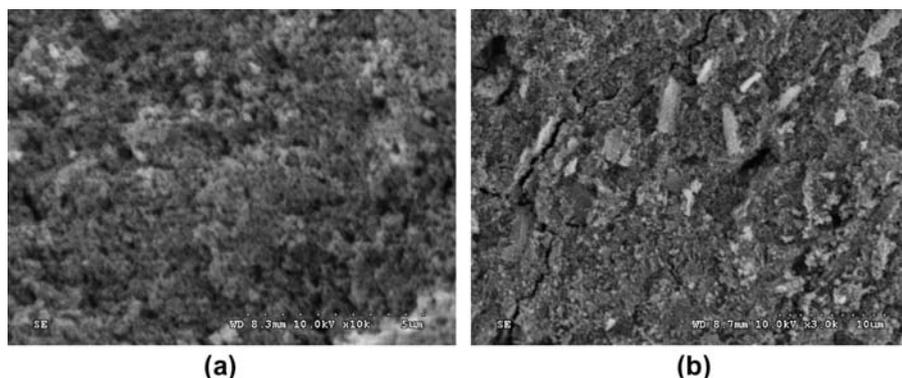


Fig. 8. Images of the flocs after the slow mixing: (a) ferrihydrite + As, 20 mg/g; (b) 50 mg/g PAC + ferrihydrite + As, 20 mg/g.

were calculated according to the method reported by Yu et al. [19]. The D_f values in different coagulation times are shown in Table 1. The mass fractal dimension D_f is restricted to the same range of Q values in order to eliminate the polydispersity effect reported by other researchers [24]. As shown in Table 1, differences were apparently in the scattering dates of ferrihydrite particles with different flocculants added. D_f value was calculated in the log Q range of -0.8 to 0.8 according to Eq. (1), the D_f values and $D(4,3)$ were shown in Table 1.

Floc size and fractal dimension of aggregated ferrihydrite in different times of the coagulation process were shown in Table 1. Floc size increased significantly during the coagulation process when PFS or PAC were used as the coagulant. The size of aggregated ferrihydrite particles with PAC added was larger than ferrihydrite particles or with PFS added. Meanwhile, the mass fractal dimension of flocs with PAC added was larger than with PFS added. The results were in accordance with the discussion in Section 3.2 that the coagulation efficiency of PAC was higher than PFS in pH 6.5. The structure of aggregated ferrihydrite particles formed by PAC added was of advantage in settling.

As the aggregate size increases over time, the fractal dimension usually decreases and the floc structures become more open. In the flocculation with PAC or PFS, however, the slope of the scatter patterns increases slightly at the intermediate scale, implying an increase in the fractal dimension. It is partly possible that the large scale flocs are joined together to form more compact and stable structures due to restructuring and rearrangement. Furthermore, the images of ferrihydrite flocs, as shown in Fig. 8, clearly indicate that the surface of flocs became more compact after PAC was added.

The floc structures by PFS are more open in comparison with those by PAC. As discussed in Section 3.2, the flocculation rates by PAC are faster compared with those in PFS. Negative charge on the surface of flocs inhibited the formation of aggregated ferrihydrite particles with PFS added, resulting in a more opened floc structure. The invariable value D_f of flocs with PFS or PAC added showed no reconstruction and no rearrangement in the coagulation process.

4. Conclusion

The coagulation of arsenic adsorbed ferrihydrite was investigated with the use of PFS or PAC. Carbonate rich ferrihydrite was prepared to simulate the flocs produced in natural aqueous arsenic treatment. Comparisons of coagulation process of ferrihydrite particles with PAC or PFS were studied. Acidic circumstance was of advantage for PFS to destabilize arsenic adsorbed ferrihydrite particles, while PAC was effective on coagulation process at neutral or base conditions. The removal efficiency of arsenic adsorbed ferrihydrite particles with PAC or PFS was significantly improved. The particle size and mass fractal dimension of flocs with PAC or PFS added were also larger than arsenic adsorbed ferrihydrite.

Acknowledgements

Financial support provided by the Special fund for technology development from research institutes supplied by the Ministry of Science and Technology (under Grant No. 2010EG111021) and Ph.D. programs foundation of East China Institute of Technology.

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