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original article

Arsenic removal by coagulation using ferric chloride and chitosan from water

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ABSTRACT

Aims: In this study, the effect of arsenite and arsenate removal in synthetic water by coagulation using ferric chloride ($FeCl_3$) as coagulant and chitosan as coagulant aid were investigated.

Materials and Methods: A conventional jar test apparatus was used to evaluate the coagulation process. The effects of different conditions of pH (5.5-9), coagulant doses of $FeCl_3$ 0–60 mg/l), Arsenic speciation (As (V) and As (III)), Chitosan as coagulant aid, and initial arsenic concentrations (0.2–2 mg/l) to Arsenic removal was investigated by considering the residual arsenic and iron after coagulation.

Results: The finding showed that the optimum pH for $FeCl_3$ as a coagulant to As (V) and As (III) removal was 7. As (V) removal efficiency of 90% or higher by $FeCl_3$ alone were achieved for initial As (V) concentration 0.2–2 mg/l at optimum dosages (15-30 mg/l). In contrast with As (V) removal, about 60% of the As (III) were removed in comparable conditions. The optimal dosage of chitosan was determined to be 0.5 mg/l. When chitosan (0.5 mg/l) was used as coagulant aid with $FeCl_3$, the efficiency removal was enhanced almost to 100% and 80% for As (V) and As (III) removal, respectively. The optimum dosage of $FeCl_3$ was reduced, especially at low initial concentration of arsenic.

Conclusions: Chitosan as natural coagulant aid improved arsenic removal efficiency by coagulation process using $FeCl_3$. This method can be used for regions with drinking water contaminated with initial arsenic concentration less than 1 000 μg/l.

Key words: Arsenic, chitosan, coagulation and flocculation, ferric chloride, natural coagulant aid

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INTRODUCTION

Arsenic (As) is the 20th most abundant element in the earth's crust and most of the people are exposed with this substance in their life.[1] Arsenic may be present either as organic or inorganic form. Inorganic arsenic generally exists in two predominant oxidation state, arsenate (H_3AsO_4, H_2AsO_4, and

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AsO_4^{3-}) and arsenite (H_3AsO_3, $H_2AsO_3^-$, and $HAsO_4^{2-}$). The predominant oxidation state depends on the pH and the redox potential. Arsenite (As (III)) toxicity is more than arsenate (As (V)).[2-5] Arsenic is a common pollutant in underground water and industry sewage.[6,7] Arsenic exposure via drinking water is a global problem and has been reported in many countries, especially Argentina, Bangladesh, India, Mexico, Canada, Japan, China, Taiwan, and some regions of Iran (Kordestan, Northern Khorasan, and Eastern Azerbayejan).[4,7,8] In Asia, chronic arsenic poisoning is turning into an emergency epidemic and more than 100 million people are exposed to arsenic contamination in water.[9] Based on epidemiologic studies, one of the most serious effects of inorganic arsenic toxicity is the arsenic carcinogenic influence on human beings; the element is classified as group one carcinogenic substance.[10] Non-organic arsenic causes skin, lung, bladder, liver, and kidney cancer and some of its non-cancer effects include skin injuries, cardiovascular diseases, hypertension, chronic cough, adverse effects of pregnancy, toe gangrene, diabetes mellitus, and neurotoxicity.[3,6,11,12] Epidemiological studies showed that about 10% of people use drinking water including 500 μg/l arsenic die due to cancer[13] and 60 mg of arsenic is fatal for human being.[14] Iranian standard of arsenic in drinking water is 0.05 mg/l,[8] while WHO and EPA have recommended a maximum contaminant level of 0.01 mg/l arsenic in drinking water.[13]

Normally, there are some methods for arsenic removal including coagulation, co-precipitation, adsorption process, ion exchange, reverse osmosis, membrane filtration, and alternative methods such as oxidation with ozone, bioremediation, and electrochemical remediation.[4] The above methods except of coagulation method although have a high efficiency in removing arsenic but due to high costs, their use are limited. Coagulation method has been widely used owing to the low cost and high efficiency for the removal of arsenic, turbidity, and color.[15] The applied coagulants are alum and ferric chloride ($FeCl_3$). Arsenic removal efficiency by $FeCl_3$ is better than alum because its ions are mostly inclined to ferrous compounds.[4,16] Baskan *et al.* found that coagulation by $FeCl_3$ with the dosage of 30 mg/l could decrease arsenic from 50 to 10 μg/l. This study showed that among ferrous compounds including $FeCl_3$, ferric sulfate, and ferrous sulfate, $FeCl_3$ had high efficiency in arsenic removal.[3] Also, Hering *et al.* found that arsenate removal by coagulation using $FeCl_3$ was efficient than arsenite removal and arsenite could not be removed by coagulation with alum.[17,18] Han *et al.* indicated that coagulation and electrocoagulation by $FeCl_3$ could remove arsenic in water effectively.[14] Complete removal of arsenate was obtained by coagulation using $FeCl_3$.[19]

Chitosan as cationic polysaccharide is an important coagulant biopolymer in water treatment and is one of the derivatives of chitin. Compared to natural coagulants, chitosan has some advantages such as low required dosage, formation of bulk flocs, high precipitation speed, biological degradation, easy sludge treatment, high efficiency in removal of suspended solids and metal ions, anti-bacterial, and without secondary pollution.[20,21] Ching-chun *et al.* showed the high efficiency of chitosan as a sorbent in arsenic removal.[20] Gupta *et al.* evaluated iron–chitosan composites could remove arsenic via absorbing in arsenic concentration from 500 μg/l to below 10 μg/l.[16]

The major objective of this study was to investigate the removal efficiencies of arsenate and arsenite from synthetic water by coagulation and flocculation using $FeCl_3$ as coagulant and chitosan as natural coagulant aid. Furthermore, optimum coagulant and coagulant aid by considering residual arsenic less than 50 μg/l and residual iron less than 0.3 mg/l (MCL OF Iran) were determined.

MATERIALS AND METHODS

Preparation of solutions

Chitosan powder (100 mg) with deacetylation degree of 85% (Aldrich) was accurately weighed into a glass beaker, mixed with 10 ml of 0.1M HCl solution, and kept aside for about one hour to dissolve. It was then diluted to 100 ml with distilled water. This solution should be prepared daily. HCl was considered to be a better choice for as acid environment compared to acetic acid to avoid the entrance of organic matter to the sample by acetic acid.[22]

A few grams weighed Kaolin (Sigma-Aldrich) was kept in the oven for 3 or 4 hours at 105°C to be dried. Then, it was taken out to be placed in desiccator to be cooled for 30 minutes. It was then diluted to 1.5 l with distilled water and stirred for 1 hour to uniform dispersion of kaolin particle. The mixture was permitted to stand for 24 hours to allow to complete hydration of the kaolin. After the surface liquid was withdrawn, it was poured into glass flask and was kept as stock solution.[22] This suspension was used to prepare turbid water with turbidity of 10 NTU.

To prepare arsenate and arsenite solutions, sodium arsenate ($Na_2HAsO_4.7H_2O$) and sodium arsenite ($NaAsO_2$) (Aldrich and Merck Co., respectively) were dissolved in distilled water in concentration of 1 000 mg/l. The solutions were prepared daily. These stock solutions were used to prepare synthetic water spiked with different arsenic concentrations in the range of 0.2, 0.5, 1, and 2 mg/l which are common in drinking water of contaminated regions of Iran.[9,23]

It was prepared by dissolution of $FeCl_3.6H_2O$ (Merck) with distilled water with the concentration of 10 000 mg/l.

Experimental procedure

A conventional jar test apparatus (Model 7790-402) was used for coagulation experiments. First, one liter of water sample in accordance with the characteristics of Table 1 was added to each beaker. Before the addition of the coagulant,

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the pH of water sample was adjusted using hydrochloric acid (HCl) and sodium hydroxide (NaOH) solutions and spiked with As (V) or As (III) by arsenic stock solutions. After the coagulant was added to the suspension, the beaker was rapidly mixed at 100 ± 2 rpm for 1 minute followed by 20 minutes of slow mixing at 40 ± 2 rpm, and afterward the samples were allowed to settle for 30 minutes. The experiments were done at environment temperature (25°C). The effect of pH changes in removal of arsenate and arsenite by $FeCl_3$ (30 mg/l) at pH 5.5-9 were investigated. Then, effect of arsenic removal by $FeCl_3$ as coagulant in various dosages (0–60 mg/l) and initial arsenic concentrations (0.2–2 mg/l) were studied. The efficiency of $FeCl_3$ as coagulant in various dosages (0–30 mg/l) with optimized chitosan as coagulant aid was studied. Chitosan as coagulant aid was added to the solution at the end of rapid mix stage. After each experiment, to determine the residual arsenic, 50 ml of sample was withdrawn from the top 2 cm of the water surface and filtered using paper filter.

Analytical methods

The residual As (V) and As (III) was determined by EZ arsenic test kit (HACH) rapidly, that its accuracy and validity has been proved in the previous studies.[24,25] The residual turbidity was determined using a turbidimeter (Eutech, TN-100). A pH-meter (CG 824) also was used for pH analysis. To analyze of residual iron after coagulation and sedimentation, atomic absorption (Perkin-Elmer model - 2380) was used. All the experiments were performed in triplicate to evaluate test reproducibility under identical conditions and all values represent the average of the triplicate experiments.

RESULTS

Based on Jar test results, the optimum pH of $FeCl_3$ as a coagulant for As (V) and As (III) removal was 7 and at the high acidic and high alkaline pH, the effect of coagulant was reduced [Figure 1]. The effect of initial arsenic concentration and oxidation state on the efficiency of arsenic removal during coagulation with $FeCl_3$ (0–60 mg/l) at pH 7 was shown in Figures 2a and b. Results indicated that As (V) removal was better than As (III) removal. As (V) removal efficiency of 90% or higher were achieved, especially for low initial As (V) concentration at optimum dosage of $FeCl_3$. In contrast, about 60% of the As (III) was removed in comparable conditions. Optimum dosage of $FeCl_3$ as coagulant without addition of chitosan by considering residual As (V) and iron for As (V) removal in initial concentration 0.2, 0.5, 1, and 2 mg/l was obtained to be 15, 25, 30, and 30 mg/l, respectively. Although at

Table 1: The water quality characteristics used for the experiments

Parameter	Concentration	
	Mean	SD
pH	8.4	-
Turbidity (NTU)	0.6	0.2
soluble solids (mg/l)	540	0.07
Total alkalinity (mg/l)	200	17.56
Total hardness (mg/l)	240	12.06
Calcium (mg/l)	64	5.51
Magnesium (mg/l)	35	2.52
Sulfate (mg/l)	50	5.57
Chloride (mg/l)	50	3.21
Carbonate (mg/l)	0	0
Bicarbonate (mg/l)	244	14.73
Iron (mg/l)	0.09	0.05
As (mg/l)	0	0

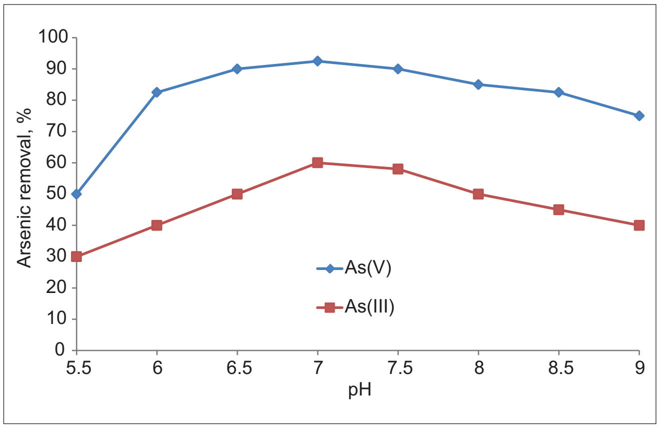

Figure 1: Effect of pH on arsenate and arsenite removal at initial arsenic concentration of 1 000 µg/l and $FeCl_3$ concentration of 30 mg/l

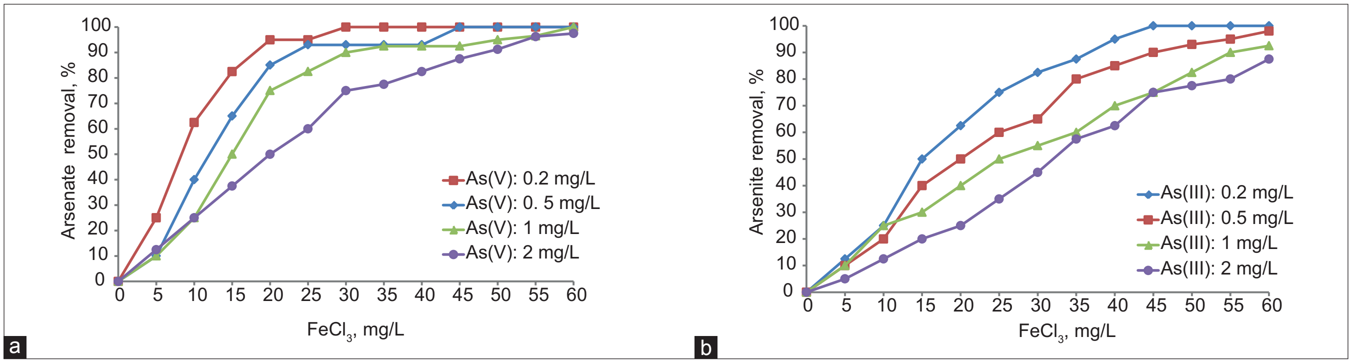

Figure 2: The effect of ferric chloride alone to removal of (a) As (V) and (b) As (III) at pH 7

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concentration higher than 30 mg/l of FeCl$_3$, the efficiency removal of As (V) was enhanced but as a shown in Figure 3, residual iron after coagulation was more than 0.3 mg/l, which is the Iranian maximum contaminant level of iron concentration in drinking water.[8] Optimum dosage of FeCl$_3$ for As (III) removal in initial concentration 0.2, 0.5, 1, and 2 mg/l was obtained to be 20, 30, 30, and 30 mg/l, respectively. Also, in this study, the optimum chitosan dosage as a coagulant aid for As (V) and As (III) removal was obtained to be 0.5 mg/l [Figure 4]. Then, the effect of FeCl$_3$ (0–30 mg/l) as a coagulant with chitosan (0.5 mg/l) as a coagulant aid on As (V) and As (III) removal is shown in Figure 5a and b. Our finding showed that efficiency removal of As (V) and As (III) was increased and optimal dosage of FeCl$_3$ was decreased. The efficiency removal of As (V) increased from 90% to 100% for low initial concentration of As (V), but complete removal was not observed for high initial concentration under the range of conditions examined. Optimal dosage of FeCl$_3$ decreased to 10, 20, and 25 mg/l for initial concentration of 0.2, 0.5, and 1 mg/l As (V), respectively, but was not decreased for initial concentration 2 mg/l As (V). The efficiency removal of As (III) increased from about 60% to 80% and optimal dosage of FeCl$_3$ was reduced from 20 and 30 mg/l to 15 and 25 mg/l for initial concentration of 0.2 and 0.5 mg/l As (III). Nevertheless, it was not decreased for initial concentration 1 and 2 mg/l As (III). As it can be seen, efficiency removal of As (V) and As (III) at low initial concentration of arsenic was more than high initial concentration of arsenic. In the above conditions, the residual turbidity was reduced below 1 NTU.

DISCUSSION

The effect of different pH (5.5-9) on arsenate and arsenite removal efficiency was investigated at an initial arsenic concentration of 1 000 μg/l and FeCl$_3$ concentration of 30 mg/l. In this study, optimum pH for arsenate and arsenite removal was obtained to be 7 and in the high acidic and high alkaline pH, the effect of coagulant was markedly reduced [Figure 1]. The effect of pH in removal efficiency of arsenic is related to solubility of amorphous hydroxides solid of ferric ions such that the flocs formed in neutral pH are more stable.[19] The results obtained from the present study were in agreement with the studies conducted by others.[2,19,26]

Our finding showed that by increase of coagulant dose, As (V) and As (III) removal efficiency was increased due to the increase of particle accumulation and formation of more flocs. As (V) removal efficiency of 90% or higher were achieved for initial As (V) concentration 0.2–2 mg/l [Figure 2]. Arsenate and arsenite anions are found in contaminated water resources as soluble. Thus, adding FeCl$_3$ as a coagulant convert soluble arsenate and arsenite anions into insoluble products and is removed in coagulation and precipitation process.[3] This products might be formed through the following three major steps: (1)

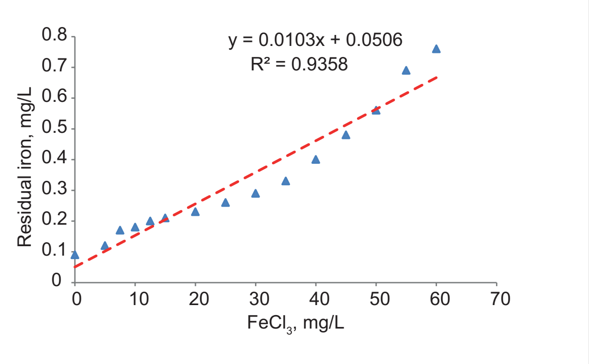

Figure 3: Residual iron after coagulation using FeCl$_3$ at pH 7

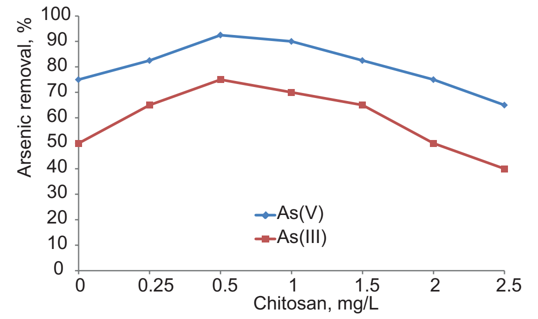

Figure 4: Determination of optimum dose of chitosan as a coagulant aid to removal of As (V) and As (III) at initial arsenic concentration of 1 000 μg/l and FeCl$_3$ concentration of 20 mg/l at pH 7

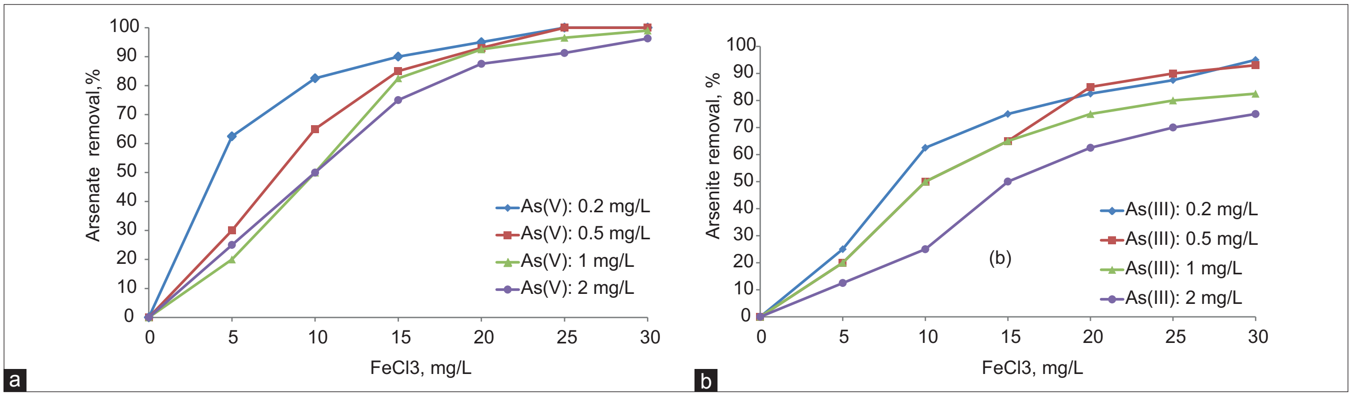

Figure 5: The effect of chitosan (0.5 mg/l) as a coagulant aid with FeCl$_3$ in the removal of (a) As (V) and (b) As (III) at pH (7)

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precipitation as solid form Fe (AsO_4), (2) Co-precipitation with iron hydroxides where arsenate and arsenite are attached to growing iron hydroxide via inclusion, occlusion, or adsorption, (3) adsorption via forming surface complexes among solute arsenic and metal solid hydroxides.[19] Donmez and Akbal investigated removal of As (V) from drinking waters by coagulation process using iron salts. Their results indicated that by increase of $FeCl_3$ dosage, the efficiency removal was increased.[19] Hering *et al*. also determined the arsenic removal from drinking water during coagulation and investigated the effect of dosage $FeCl_3$ on arsenic removal. Their findings illustrated that arsenate and arsenite removal was improved by increasing coagulant dose.[17] But excess doses of $FeCl_3$ resulted in high residual iron after coagulation and its restricted usage of high concentration of coagulant to complete arsenic removal.

Arsenic oxidation state (As (V) or As (III)) had considerable effect on arsenic removal efficiency. Our findings indicated that efficiency removal of arsenate was better than arsenite [Figure 2]. This effect is attributed to arsenate existing either as a monovalent anion, H_2AsO4^-, or as a divalent anion, $HAsO_4^{2-}$, while arsenite is found in its uncharged form, H_3AsO_3, under most pH conditions.[3] Since many removal processes including coagulation and co-precipitation, in which arsenic becomes bound or adsorbed on coagulant flocs, depend on the anionic nature of arsenic species, arsenite is much less readily removed. For this reason, oxidation of arsenite to arsenate may be necessary in order to effectively remove arsenic from water sources that contain high concentrations of arsenite. The results are in agreement with the studies conducted by Hering *et al*. and Ashraf Ali *et al*.[2,17]

In this study, the effect of chitosan as a natural coagulant aid, with $FeCl_3$ as a coagulant on arsenic removal efficiency was studied. The optimum dosage of chitosan was achieved to be 0.5 mg/l. The efficiency removal of As (V) increased from 90% to 100% for low initial concentration of As (V), but complete removal was not observed for high initial concentration under the range of conditions examined. Also, the efficiency removal of As (III) increased from about 60% to 80%. Generally, chitosan improved coagulation process and formed heavier and bigger flocs and the settlement speed of coagulation were increased. It can be said that chitosan as a coagulant aid reduced $FeCl_3$ consumption. Based on the studies conducted by Roussy *et al*., prevalent mechanism of removal of anions by chitosan as cationic polysaccharide (due to high amount of amine groups and production of high positive charge in neutral and a little acidic condition) was charge neutralization and bridging.[27] In higher concentrations of chitosan, reduction of arsenic removal was observed. It seems that high increase of positive charges of chitosan with $FeCl_3$ caused negative effect on coagulation process and re-stability of anions and subsequently the arsenic removal efficiency was reduced. There were no studies about use of chitosan as coagulant aid with $FeCl_3$ and in most of the studies, chitosan was used as sorbent material alone or with combination of other materials for arsenic removal.[16,20] Ching-chun *et al*. showed that chitosan as a sorbent in arsenic removal had a high efficiency.[20] Gupta *et al*. evaluated iron–chitosan composites via absorbing that reduced arsenic concentration from 500 μg/l to below 10 μg/l.[16]

The results of the experiments showed that by common water treatment method (coagulation and precipitation) using $FeCl_3$ as a coagulant and chitosan as a natural coagulant aid, the arsenic of water reduced to below the maximum contaminant level of Iran (50 μg/l) at low initial As (V) and As (III) and chitosan improved the coagulation process. This method can be used for regions with drinking water contaminated with average arsenic (less than 1 000 μg/l) and there is no need to complex and costly methods such as reverse osmosis and ion exchange. But this method could not decrease high initial concentration of arsenic to below maximum contaminant level of Iran.

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