# The Removal of As(V) from Drinking Waters by Coagulation Process using Iron Salts

M. Donmez, F. Akbal

*Abstract*—In this study arsenate [As(V)] removal from drinking water by coagulation process was investigated. Ferric chloride ($FeCl_3.6H_2O$) and ferrous sulfate ($FeSO_4.7H_2O$) were used as coagulant. The effects of major operating variables such as coagulant dose (1–30 mg/L) and pH (5.5–9.5) were investigated. Ferric chloride and ferrous sulfate were found as effective and reliable coagulant due to required dose, residual arsenate and coagulant concentration. Optimum pH values for maximum arsenate removal for ferrous sulfate and ferric chloride were found as 8 and 7.5. The arsenate removal efficiency decreased at neutral and acidic pH values for Fe(II) and at the high acidic and high alkaline pH for Fe(III). It was found that the increase of coagulant dose caused a substantial increase in the arsenate removal. But above a certain ferric chloride and ferrous sulfate dosage, the increase in arsenate removal was not significant. Ferric chloride and ferrous sulfate dose above 8 mg/L slightly increased arsenate removal.

*Keywords*—Arsenic removal, Coagulation, Iron salts, Drinking water

## I. INTRODUCTION

ARSENIC is a ubiquitous element, which occurs naturally in the earth's crust. Both inorganic and organic forms of arsenic have been determined in water [1]. The major arsenic species present in natural waters are arsenate ions: $H_3AsO_4$, $H_2AsO_4^-$, $HAsO_4^{2-}$, and $AsO_4^{3-}$ (oxidation state V) and arsenite ions, $H_3AsO_3$, $H_2AsO_3^-$ and $HAsO_3^{2-}$ (oxidation state III). However, As(V) ions are most prevalent in oxygenated water while As(III) is found in anaerobic conditions like in well water or in groundwater [2]. The concentration of arsenic species is mainly dependent on redox potentials. Arsenic contamination of the ground water occurs by both natural processes such as weathering of arsenic containing minerals and anthropogenic activities such as uncontrolled industrial discharge from mining and metallurgical industries, and application of organoarsenical pesticides. Inorganic arsenic is predominantly present in natural waters [3]. Arsenic cannot be easily destroyed and can only be converted into different forms or transformed into insoluble compounds in combination with other elements, such as iron [4]. The presence of arsenic compounds in groundwater, one of the main sources of drinking water, is a serious environmental and health problem. Increased concentrations of arsenic in natural waters have been reported in many regions of the world [5].

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Due to its high toxic effects on human health, in the US, the maximum contaminant level (MCL) in drinking water used to be 50 µg/L. However, the US Environmental Protection Agency (USEPA) recently finalized a new MCL of 10 µg/L. This new MCL is the same as stated by the World Health Organization guidelines [6].

The lowering of As drinking water standard requires effective and cheap technologies for As removal from the As drinking water. Among a variety of technologies (including precipitation coagulation, membrane separation, ion exchange, lime softening and adsorption), adsorption and coagulation are believed to be the cheapest As removal methods [7]. Some recent treatment technologies based on oxidation and adsorption are, iron oxide coated sand [8]-[9], manganese dioxide coated sand [10], clay minerals [11] and zero-valent iron [12]. The USEPA has identified seven technologies as the best available technologies (BATs), which are given in Table 1. Coagulation is included among these BATs [6].

This study investigates the feasibility of the ferrous and ferric ions for As(V) removal from drinking water. The main objectives are (i) to evaluate the impact of coagulant dose and pH (ii) to determine the As(V) removal efficiency.

## II. EXPERIMENTAL METHODS

TABLE I
BEST AVAILABLE TECHNOLOGIES AND THEIR ARSENIC REMOVAL EFFICIENCIES

| Treatment Technologies | Maximum removal, % |
|---|---|
| Ion exchange (sulfate 50 mg/L) | 95 |
| Activated alumina | 95 |
| Reverse osmosis | >95 |
| Modified coagulation/filtration | 95 |
| Modified lime softening (pH>10.5) | 90 |
| Electrodialysis reversal | 85 |
| Oxidation/filtration (iron:arsenic=20:1) | 80 |

Percentage removal values are for arsenat removal. Preoxidation may be required to convert arsenit to arsenat

### A. Materials

All chemicals used were of analytical grade. All glass and plastic equipment used in bench-scale testing was cleaned and acid washed using %10 nitric acid, triple-rinsed with deionized water. Stock solution of arsenate containing 1000 mg As/L was prepared using $Na_2HAsO_4.7H_2O$ and was stored at 4°C in the refrigerator. An arsenate solution at an initial experimental concentration of 100 µg As/L was prepared on the day of experimentation by diluting the stock solution with tap water. The chemical composition and some properties of the tap water used in this study are listed in Table 2.

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Solutions of Fe(II) and Fe(III) at 1000 mg/L concentrations were also prepared fresh using $FeSO_4.7H_2O$ and $FeCl_3.6H_2O$ salts, respectively. The solutions were prepared with deionised water. Hydrochloric acid and sodium hydroxide solutions were prepared for pH adjustment.

TABLE II
CHARACTERIZATION OF TAP WATER USED FOR EXPERIMENTS

| Components | Values |
|---|---|
| pH | 7.26 |
| Conductivity (µS/cm) | 265.02 |
| Temperature (°C) | 18.5 |
| Turbidity (NTU) | 0.5 |
| Aluminum (mg/L) | 0.076 |
| Calcium (mg/L) | 44.67 |
| Chloride (mg/L) | 5.9 |
| Magnesium (mg/L) | 8.3 |
| Manganese (mg/L) | 0.0097 |
| Sodium (mg/L) | 7.53 |
| Nitrate (mg/L) | 2.77 |
| Potassium (mg/L) | 0.039 |
| Sülfate (mg/L) | 16.09 |
| T. Iron (mg/L) | 1.75 |

*B. Experimental procedure*

Batch-scale experiments were conducted by transferring 200 mL of 100 µg As(V)/L arsenate solutions to 500 mL jar . Experiments were conducted using the standard jar test apparatus. Coagulation was carried out with ferrous sulfate and ferric sulfate. The coagulant was added to jar containing the sample water. The samples were mixed with rapid mixing at 120 rpm. After 1 min of rapid mix, 30 min of slow mixing at 40 rpm was provided, followed by at 30 min of settling. At the end of the settling period, water samples were taken from the supernatants, immediately vacuum filtered using a 0.45 µm pore size membrane filter, and stored at 4ºC for further analysis. Prior to addition of coagulant, the sample water pH was adjusted by adding HCl or NaOH.

*C. Arsenic analysis*

Arsenic in the influent and effluent aqueous solutions was measured by the hydride generation procedure coupled with ICP-atomic emission spectrometry.

## III. RESULTS AND DISCUSSIONS

*A. Effect of pH*

The effect of pH on arsenate removal efficiency was investgated at an initial arsenate concentration of 100 µg/L. Fig. 1 and 2 show the residual arsenate concentrations and arsenate removal efficiencies at different pH and a constant initial arsenate concentration of 100 µg/L with the ferrous sulfate and ferric chloride concentration of 10 mg/L The arsenate removal increased with increasing pH up to nearly 8 and 7.5 for ferrous sulfate and ferric chloride, respectively. As it can be seen, in the high acidic and high alkaline pH for Fe(III) caused lower efficiencies. However, Fe(II) was effective at pH 8 and form ferrous hydroxide in the alkaline range. The arsenate removal efficiency decreased at neutral and acidic pH values for Fe(II).

The effect of the pH on the arsenate removal efficiency is related to the solubility of the amorphous hydroxide solid of ferric ions. Arsenate removal efficiency with Fe(III) increased in the pH range of 6 to 8 because of the fact that the amorphous hydroxide solid is stable in this pH range. Obtained results at optimum pH values of coagulants are in good agreement with the values given in literatures [6]-[13]-[14].

*B. Effects of coagulant dose*

Fig. 3 and 4 show the variations of residual arsenate concentrations and arsenate removal effciencies with initial arsenate concentration of 100 µg/L at different $FeSO_4$ and $FeCl_3$ concentrations respectively. The addition of coagulant caused a substantial increase in the arsenate removal. But above Fe(II) and Fe(III) dose above 8 mg/L the increase in arsenate removal was not significant.
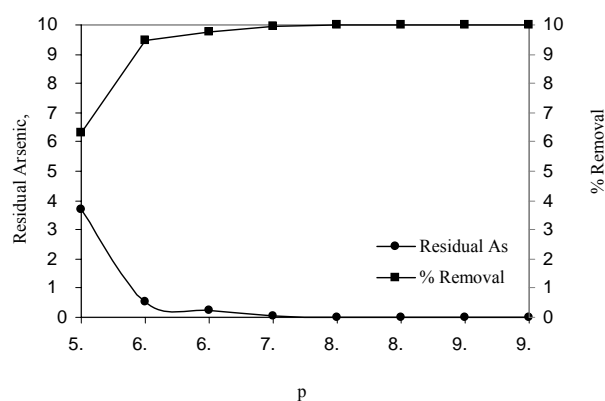


Fig. 1 Effect of pH on arsenate removal at initial arsenate concentration of 100 µg/L and ferrous sulfate concentration of 10 mg/L
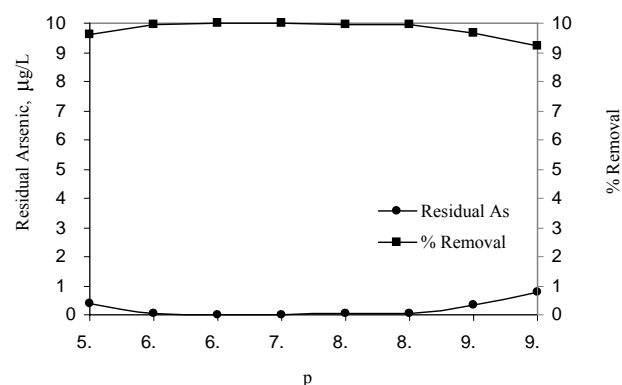


Fig. 2 Effect of pH on arsenate removal at initial arsenate concentration of 100 µg/L and ferric chloride concentration of 10 mg/L

World Academy of Science, Engineering and Technology
International Journal of Environmental and Ecological Engineering
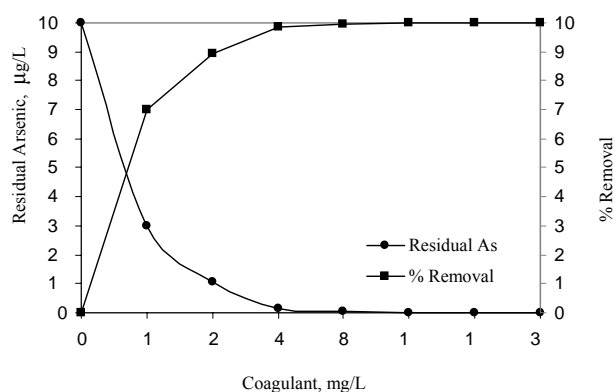Vol:5, No:6, 2011

Fig. 3 Effect of coagulant dose on arsenate removal at initial arsenate concentration 100 μg/L, constant pH of 8.0 and different ferrous sulfate concentrations
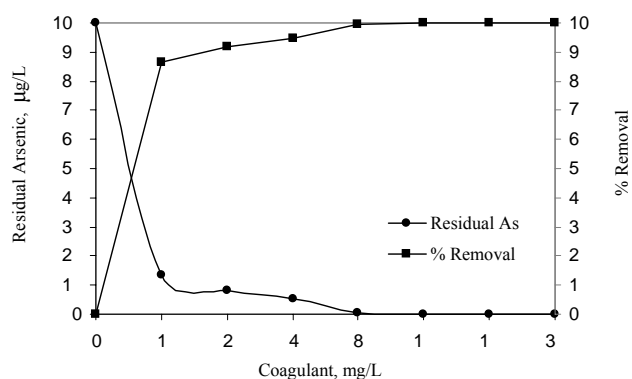


Fig. 4 Effect of coagulant dose on arsenate removal at initial arsenate concentration of 100 μg/L, constant pH of 7.5 and different ferric chloride concentrations

Ferric chloride was more effective and efficient than ferrous sulfate in reducing arsenite concentration from 100 μg As(V)/L to <10 μg As(V)/L. As(V) is a negatively-charged anion and sorbs to the positively-charged $Fe(OH)_3$ particles or flocs.

Pentavalent arsenic exists in anionic form of $H_2AsO_4^{4-}$, $HAsO_4^{2-}$ or $AsO_4^{3-}$ above the pH of 2. Thus, addition of iron coagulants to water could facilitate the conversion of soluble arsenic species to insoluble reaction products. These products might be formed through three major steps: (i) precipitation in forms of $Fe(AsO_4)$ solid; (ii) coprecipitation where soluble arsenic species were incorporated into a growing hydroxide phase via inclusion, occlusion, or adsorption; and (iii) adsorption involving the formation of surface complexes between soluble arsenic and the solid hydroxide surface site [15].

## IV. CONCLUSIONS

Coagulation process was used for arsenate (As(V)) removal from drinking water. Ferric chloride were used as source of Fe(III) and ferrous sulfate was used as source of Fe(II). Coagulant dose and pH were chosen as independent variables. Experimental results demonstrated that the Fe(III) ions are more effective for arsenate removal. Optimum pH values for maximum arsenate removal for ferrous sulfate and ferric chloride were found as 8 and 7.5. The arsenate removal increased with increasing coagulant concentration as a result of increasing amount of coagulant with increasing rate of particle aggregation and floc formation. For the initial arsenate concentration of 100 μg/L, the arsenate removal efficiency was obtained 100% at the $FeSO_4$ and $FeCl_3$ concentrations of 10 mg/L.

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