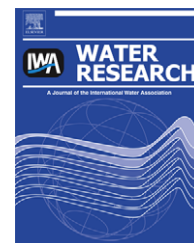


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# Removal of arsenic from water: Effects of competing anions on As(III) removal in $\text{KMnO}_4$ -Fe(II) process

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## ABSTRACT

Effects of sulfate, phosphate, silicate and humic acid (HA) on the removal of As(III) in the  $\text{KMnO}_4$ -Fe(II) process were investigated in the pH range of 4–9 with permanganate and ferrous sulfate applied at selected dosage. Sulfate decreased the removal of arsenic by 6.5–36.0% at pH 6–9 and the decrease in adsorption did not increase with increasing concentration of sulfate from 50 to 100 mg/L. In the presence of 1 mg/L phosphate, arsenic removal decreased gradually as pH increased from 4 to 6, and a sharp drop occurred at pH 7–9. The presence of 10 mg/L silicate had negligible effect on arsenic removal at pH 4–5 whereas decreased the arsenic removal at pH 6–9 and the decrease was more significant at higher pH. The presence of HA dramatically decreased the arsenic removal over the pH range of 6–9 and HA of higher concentration resulted in greater drop in arsenic removal. The effects of the competing anions on arsenic removal in the  $\text{KMnO}_4$ -Fe(II) process were highly dependent on pH and the degree of these four anions influencing As(III) removal decreased in the following order, phosphate > humic acid > silicate > sulfate. Sulfate differed from the other three anions because sulfate decreased the removal of arsenic mainly by competitive adsorption while phosphate, silicate and HA decreased the removal of As(III) by competitive adsorption and sequestering the formation of ferric hydroxide derived from Fe(II).

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## 1. Introduction

Arsenate (As(V)) and arsenite (As(III)) are common arsenic species in naturally contaminated groundwater and surface water in many countries. Arsenic has been considered as a strong poisonous chemical due to its odorless and nearly tasteless nature (Jeong and Fan, 2007). Even small amounts of arsenic in drinking water have adverse effects on human health. Arsenic can be concentrated in liver, lung, kidney, bladder, and skin by the ingestion of dissolved arsenic (Wu et al., 1989). Due to its high toxicity, the United States Environmental Protection Agency (USEPA), the World Health Organization (WHO) and the Ministry of Health of P.R. China

have decreased the maximum contaminant level (MCL) of arsenic in drinking water from 50 to 10  $\mu\text{g/L}$  (Guan et al., 2009b). However, elevated arsenic concentrations (>50  $\mu\text{g/L}$ ) have often been found in many groundwaters around the world. In some cases, the concentration can reach a level of 1–10 mg/L (Lien and Wilkin, 2005). The discrepancy between the high arsenic concentrations in raw water and the stringent standard calls for an emergent modification of the current treatment technology and a development of new treatment technologies.

Arsenic exists in water primarily as the inorganic oxyanions of arsenite (As(III)) and arsenate (As(V)), with As(III) predominating in anaerobic waters and As(V) prevailing in oxic waters

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(Liu et al., 2008). Recent surveys about arsenic redox speciation in groundwater, which is a major source of drinking water throughout the world (Smedley et al., 2002), suggest that As(III) can represent up to 67–99% of total arsenic in groundwater (Mukherjee and Bhattacharya, 2001). Since arsenite is more mobile and toxic than arsenate, and As(III) is predominant in many groundwaters, active remediation of As is often required that may involve conversion of As(III) to As(V), and immobilization of As(V) by adsorption or coprecipitation.

Lee et al. (2003) employed ferrate to oxidize As(III) to As(V) and then remove As(V) by Fe(III) coagulation. Liu (2005) applied potassium permanganate to oxidize As(III) to enhance the removal of As(III) by ferric chloride. Hug et al. (2001) and Roberts et al. (2004) examined the feasibility of removing As(III) by Fe(II) with the presence of oxygen. A novel process,  $\text{KMnO}_4$ -Fe(II) process, was developed in our previous research for As(III) removal and the removal efficiency of As(III) in this process was compared with those in other oxidation-coagulation processes reported in the literature (Guan et al., 2009a). It was found that at the optimum dosage of permanganate, the  $\text{KMnO}_4$ -Fe(II) process was much more efficient than the  $\text{KMnO}_4$ -Fe(III) process for As(III) removal by 15–38% at pH 5–9. Considering the availability, stability and price of the oxidant, As(III) removal efficiency and the production of toxic sludge, it is reasonable to conclude that  $\text{KMnO}_4$ -Fe(II) is a very appealing process for As(III) removal (Guan et al., 2009a).

Arsenic is not the only inorganic ion present in natural waters and adsorption of As(V) and As(III) oxyanions by ferric hydroxide may be adversely affected by anions such as carbonate, sulfate, phosphate, silicate and natural organic matter (Meng et al., 2000). Anions directly compete for available surface binding sites and indirectly influence adsorption by alteration of the electrostatic charge at the solid surface (Jain and Loeppert, 2000). Both direct and indirect effects are influenced by solution pH, the relative anions concentrations, and intrinsic binding affinities (Mesuere and Fish, 1992). Many studies have investigated the effects of competing ions on arsenic adsorption (Wilkie and Hering, 1996; Jain and Loeppert, 2000; Jackson and Miller, 2000; Su and Puls, 2001; Arai and Sparks, 2004; Radu et al., 2005), but the results are inconsistent stating the effect for the same competing ion. The most abundant competing ligands naturally present are organic anions, hydroxyl, bicarbonate, nitrate, silicate, sulfate, and phosphate (Parfitt, 1978). Among these anions, it is believed that carbonate anions do compete with As for adsorption sites but the competitive effect is relatively small with regard to the potential competitive effects of other anions (Meng et al., 2000; Radu et al., 2005). Therefore, the effects of sulfate, phosphate, silicate and humic acid on As(III) removal in the  $\text{KMnO}_4$ -Fe(II) process were examined in this study as functions of pH and concentrations of anions. This research aimed at providing valuable information for applying the  $\text{KMnO}_4$ -Fe(II) process in the removal of As(III) in water treatment industry.

## 2. Materials and methods

All chemicals used in the experiments were reagent grade. All solutions were prepared with distilled water.  $\text{NaAsO}_2$  was

dissolved in DI water to prepare primary stock solution containing 1000 mg/L As(III) every week. Ferrous sulfate solution was prepared fresh for each set of experiments by dissolving  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  in distilled water to avoid its oxidation by air.  $\text{Na}_2\text{SO}_4$ ,  $\text{Na}_2\text{SiO}_3 \cdot 7\text{H}_2\text{O}$ ,  $\text{NaH}_2\text{PO}_4$  solutions containing 1000 mg/L  $\text{SO}_4^{2-}$ , Si and P, respectively, were prepared every week. The stock solution of humic acid was prepared by dissolving 1 g reagent grade humic acid powder into 500 ml DI water. The pH of this solution was adjusted to 12 with 1 mol/L sodium hydroxide and then the solution was filtered through a cellulose acetate membrane (MFS) of 0.45  $\mu\text{m}$  pore size. The supernatant was collected and adjusted to neutral with hydrochloric acid and transferred to a plastic bottle for storage. Experimental solutions of the desired humic acid concentrations were obtained by successive dilutions.

Jar tests were performed with a standard jar testing device (Stuart Scientific) to simulate a conventional coagulation/flocculation process to study the effects of sulfate, phosphate, silicate, or humic acid on removal of As(III) in the  $\text{KMnO}_4$ -Fe(II) process. The initial As(III) concentration was 13.3  $\mu\text{mol/L}$ , the dosage of  $\text{KMnO}_4$  was 18.6  $\mu\text{mol/L}$  in  $\text{KMnO}_4$ -Fe(II) process, and the coagulant as Fe(II) was applied at 44.6  $\mu\text{mol/L}$ . If it was not otherwise specified, the procedure of jar test was the same as described in our previous study (Guan et al., 2009a). In brief, all permanganate was dosed to the As(III)-bearing solution at the beginning of jar test (single dosing) and ferrous sulfate was applied 2 min later. In addition, in the experiments examining the effects of humic acid on As(III) removal, another permanganate dosing mode, sequential dosing (9.3  $\mu\text{mol/L}$  permanganate was applied at the beginning of jar test and 9.3  $\mu\text{mol/L}$  permanganate was applied 2 min later together with ferrous sulfate) was also employed to investigate the consumption of permanganate by humic acid.

All jar tests were carried out in a temperature controlled room at 20–23 °C and each experiment was carried out in (at least) duplicate. All experiments were performed with a constant ionic strength of 0.01 mol/L  $\text{NaNO}_3$ . Previous study revealed that alkalinity had negligible effect on arsenate removal over a wide pH range (Meng et al., 2000), thus the presence of alkalinity will not interfere with the observation of the influence of competing anions. Moreover, alkalinity is ubiquitous in natural waters. Therefore, 0.001 mol/L  $\text{NaHCO}_3$  was added to provide alkalinity in this study. All experiments were carried out open to the atmosphere. Sodium hydroxide and hydrochloric acid were used to adjust the pH of the solutions, which was kept constant throughout the jar tests. All glassware was cleaned by soaking in 10%  $\text{HNO}_3$  and rinsed three times with distilled water.

After each test, the supernatant was sampled and filtered immediately through a cellulose acetate membrane (MFS) of 0.45  $\mu\text{m}$  pore size for the determination of total As, Fe and Mn by inductively coupled plasma mass spectrometry (ICP-MS) method. In the ICP-MS method, all samples and standards were acidified according to the Standard Methods (APHA, 1995). A high performance pH meter with a saturated KCl solution as electrolyte (Corning 350) was used to measure solution pH. Daily calibration with proper buffer solutions (pH 4.00, 6.86 and 9.18) was performed to ensure its accuracy.

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