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Enhanced arsenite removal through surface-catalyzed oxidative coagulation treatment

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HIGHLIGHTS

- The addition of Fe(II) or H₂O₂ during Fe(III)-based conventional coagulation was evaluated for enhanced As(III) removal.
- H₂O₂-amended coagulation can significantly enhance As(III) removal than the conventional or Fe(II)-amended coagulation systems.
- H₂O₂-amended coagulation holds a reliable As(III) removal performance over pH 4–9.
- Among common background solutes examined, phosphate poses the greatest impact on As(III) removal during H₂O₂-amended coagulation.
- Overall, addition of a small dose of H₂O₂ can be a viable method to enhance As(III) removal in drinking water treatment processes.

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ABSTRACT

Arsenic being a naturally-occurring groundwater contaminant is subject to stringent water quality regulations. Coagulation and adsorption are widely used methods to treat arsenic-contaminated water, however, these treatments have been reported to be less efficient for the removal of arsenite (As(III)) than arsenate (As(V)). In this study, the feasibility of *in situ* oxidation of As(III) during coagulation was investigated in two systems: Fe(II) or H₂O₂-assisted oxidative coagulation treatment using ferric chloride as the coagulant. This setup exploits the catalytic property of the fresh formed Fe(III) hydroxide colloids in coagulation suspension to mediate the production of reactive oxidants capable of As(III) oxidation. Fe(II)-assisted coagulation brought about small improvements in As(III) removal compared to treatment with Fe(III) coagulant alone, however, its arsenic removal efficiency is strongly dependent on pH (observed optimal pH = 7–9). Addition of H₂O₂ together with ferric chloride led to a significant enhancement in arsenic retention at pH 6–8, with final arsenic concentrations well below the U.S.EPA regulatory limit (10 µg/L). H₂O₂-assisted oxidative coagulation can attain reliable As(III) removal over a broad pH range of 4–9. Radical quenching experiments reveal the participation of superoxide radical in As(III) removal in the oxidative coagulation systems. Phosphate (at > 0.1 mM) strongly suppresses As(III) removal efficiency, whereas carbonate and humic acid pose a minor impact. Overall, the results suggest that a low dose addition of H₂O₂ along with ferric coagulant is a feasible method for the existing water treatment facilities to achieve improved As(III) removal efficiency.

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1. Introduction

Arsenic is a contaminant of wide-spread occurrence in groundwater at diverse geological locations, and the increasing dependence of global drinking water supply on groundwater has resulted in major epidemics of arsenic poisoning in the past few decades (Khandaker et al., 2009; Smedley and Kinniburgh, 2002).

Although the use of arsenic as agricultural chemicals (e.g., insecticides and wood preservatives) has been banned in many countries (Vaughan, 2006), natural processes such as mineral weathering contribute a major source of arsenic in groundwater. In some cases, the release of arsenic from mineral deposits is aggravated by anthropogenic activities that can lead to substantial changes in subsurface biogeochemical conditions (Neumann et al., 2010; Dittmar et al., 2010).

The dominant forms of arsenic found in groundwater are arsenite (As(III), as H₃AsO₃ at neutral pH) and arsenate (As(V), as HAsO₄²⁻ at neutral pH) (Johnston and Singer, 2007; Bang et al., 2005). While

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As(III) is considered more toxic than As(V), the health risk associated with arsenic exposure also depends on the mobility of different arsenic species (Vaughan, 2006). The adsorption behavior of As(III) and As(V) onto various forms of amorphous or crystalline iron oxides has been investigated under a broad range of aqueous conditions, and iron minerals are also widely applied as adsorbents for treating arsenic-contaminated water (Mohan and Pittman, 2007; Bissen and Frimmel, 2003). Within an environmentally relevant pH range (pH 4–10), the adsorption edge of arsenite onto common iron oxides (e.g., goethite and ferrihydrite) exhibit maximum absorption over a broad pH, whereas that of arsenate declines substantially with increasing pH (Goldberg and Johnston, 2001; Dixit and Hering, 2003; Raven et al., 1998). In addition, the capacity of arsenic uptake is affected by the initial arsenic concentration, the As to Fe ratio, and the composition of background solutes (Goldberg and Johnston, 2001; Dixit and Hering, 2003). Common background anions such as phosphate, sulfate, and carbonate pose competition against arsenic species for the surface sites of iron oxides, as a result, their presence can severely impact the efficiency of arsenic adsorbents (Dixit and Hering, 2003; Su and Puls, 2001; Hug et al., 2008).

Many studies have examined the removal of arsenic from water during the conventional coagulation process. Coagulation is an essential process in a drinking water treatment system for the removal of turbidity. The treatment typically entails the addition of ferric or aluminum salts (i.e., the coagulants) into feed water during a rapid mixing step, followed by a period of slow agitation to induce flocculation of colloidal particles into large aggregates due to either charge neutralization by hydrolyzed ferric or aluminum complexes or enmeshment of small particles in polymerized floc network (Edzwald, 1993). It is considered that arsenic can be sequestered from aqueous phase during conventional coagulation via adsorption onto the floc surface and/or co-precipitating with the coagulants (Edwards, 1994; Hering et al., 1996; Meng et al., 2000). It has been shown that As(V) is more effectively removed than As(III) at neutral pH due to the sensitivity of As(III) removal to the type of coagulant used and the background constituents (Hering et al., 1996; 1997). Attempts have been made, therefore, to incorporate an oxidation step prior to arsenic removal by coagulation or adsorption. Accordingly, oxidative pretreatment using chlorine (Pio et al., 2015), permanganate (Sen et al., 2010; Bordoloi et al., 2013), or ferrate (Lee et al., 2003) have been proposed. These techniques require the addition of strong oxidants to drive homogeneous oxidation of As(III) in the aqueous phase. As(III) oxidation catalyzed by solid surfaces may present an alternative and potentially more efficient route of As(III) oxidation. Surface-mediated oxidation has been shown for various iron-containing minerals including magnetite, goethite and ferrihydrite (Ona-Nguema et al., 2010; Voegelin and Hug, 2003; Amstaetter et al., 2010; Bhandari et al., 2011). In the case of solids containing only Fe(III), it entails the addition of Fe(II) (Ona-Nguema et al., 2010; Amstaetter et al., 2010) or H₂O₂ (Voegelin and Hug, 2003) or exposure to sunlight irradiation (Bhandari et al., 2011, 2012) to generate reactive oxidants capable of As(V) formation. Moreover, As(III) oxidation in Fe(II) or H₂O₂–Fe(III) systems was not significantly affected by hydroxyl radical quenching agents at neutral pH (Hug and Leupin, 2003), suggesting the oxidants arising in these systems are fairly selective towards As(III). Considering this and the high affinity of As(III) for iron oxides, surface-catalyzed oxidation may be more advantageous for arsenite sequestration than solution-phase oxidation methods. Indeed, the concept of catalytic As(III) oxidation in the presence of iron-containing solids has been explored in recent studies in the form of photo-reduction of Fe(III) (Hug et al., 2001), electro-coagulation (Lakshmanan et al., 2010; Kumar et al., 2004), or chemically enhanced As(III) oxidation (via addition of H₂O₂)

(Hug and Leupin, 2003). However, these studies are mostly catered to point-of-use treatment systems, and the feasibility of utilizing solids generated during conventional coagulation treatment as heterogeneous catalysts for enhanced arsenic removal is a concept not explored to date. Importantly, coagulation generates a vast amount of Fe(III) hydroxide colloids and the process operates with ample hydraulic residence time, rendering coagulation basin an interesting arena to explore the reactivity of interfacial redox reactions for contaminant abatement.

The objective of this study is to evaluate the performance of two oxidative coagulation systems in treating As(III)-contaminated water. In reference to conventional coagulation using ferric chloride as the coagulant, oxidative coagulation involves the addition of a small dose of Fe(II) or H₂O₂ in addition to the coagulant to promote the formation of reactive oxidants. The effects of chemical dose, pH, and background solutes on the total arsenic removal (initially present as As(III) in the feed water) were systematically assessed. The role of As(III) oxidation in assisting its removal from the aqueous phase was investigated through comparison of the kinetics and final arsenic retention in the oxidative coagulation treatments against those in conventional coagulation as well as analysis of arsenic speciation in the solid residues of coagulation using X-ray photoelectron spectroscopy (XPS). In addition, radical quenching experiments were conducted to understand the nature of the oxidants responsible for As(III) oxidation. Finally, the performance of the proposed oxidative coagulation treatment in source water of complex composition was evaluated by examining the effects of background solutes.

2. Materials and methods

2.1. Reagents

All aqueous solutions used in this study were prepared with Milli-Q purified water (18 MΩ cm). Ferric chloride hexahydrate (FeCl₃·6H₂O), ferrous sulfate heptahydrate (FeSO₄·7H₂O), bentonite (sodium form), sodium bicarbonate (NaHCO₃), hydrogen peroxide (H₂O₂, 3% w/w) and 2-propanol (HPLC grade) were purchased from Fisher Scientific. Aluminum sulfate, commonly referred to as alum, (Al₂(SO₄)₃·14–18 H₂O) was from MCB Reagents. Sodium (meta)arsenite (NaAsO₂, 98%) and humic acid sodium salt (technical grade) were from Sigma–Aldrich. Potassium phosphate (monobasic, KH₂PO₄) and 1,4-benzoquinone were from Acros Organics. All chemicals are of ACS reagent grade or higher unless otherwise noted. FeCl₃ stock (4 g/L) was prepared daily immediately before the experiments. Arsenite stock (1000 mg/L) was also prepared fresh before each experiment.

2.2. Coagulation experiments

2.2.1. Conventional coagulation tests

All coagulation tests were performed with a 6-station floc tester (Orbeco Hellige ET750, NovaTech). The jars were 1000-mL HDPE graduated receptacles (Capitol Scientific). Synthetic feed water containing 2 mM NaHCO₃ and 0.1 g/L of bentonite to mimic turbidity in natural water was used throughout this study. Other background constituents (e.g., phosphate and humic acid) were not amended except for those experiments investigating the effect of background solutes on the coagulation performance. The feed water was stirred thoroughly and allowed to settle overnight (for approx. 16 h) prior to the experiments. Only the top portion of the feed water with turbidity of approximately 20 NTU was used. The initial pH of the feed water was adjusted to 8.0 using dilute HCl or NaOH, except for a subset of experiments evaluating the effect of pH. Immediately before the test, the feed water was spiked with

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