

北門嶼足有情—飲用水中砷去除之研究

Arsenic Removal by Coagulation with Alum

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ABSTRACT

Bench-scale studies were conducted with synthetic freshwaters to assess the applicability of coagulation processes to meet a lower standard for arsenic in drinking water. The parameters examined for their effects on arsenic removal include initial arsenic concentration and oxidation state, pH, coagulant dose.

Studies conducted in synthetic freshwaters indicated that arsenic removal by coagulation with alum was generally consistent with removal being governed by adsorption onto freshly-precipitated amorphous aluminum hydroxides. This hypothesis is supported by observations that As(V) removal by alum was more efficient than As(III) removal and that, for both As(III) and As(V), removal was independent of initial As concentration (over a limited range) and increased with coagulant dose. In addition, the negligible As (III) removal by alum, as compared with moderate removal by ferric chloride, suggested adsorption alone could not account for observed removal.

Under optimal pH conditions, however, excellent removal of As (V) could be achieved with alum, which suggests that, for influent As(V) concentrations typical of U.S. source waters, residual, dissolved As (V) concentrations $< 2 \mu\text{g/L}$ in product water can be achieved by coagulation.

Keyword: arsenic removal, adsorption, freshwaters, blackfoot disease

1. Introduction

Studies suggest that cancer risks may be significantly increased by ingestion of inorganic soluble arsenic (Chen et al., 1992; Smith et al.,

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1992). Cancer risk is associated primarily with skin cancer, though other cancers (such as liver, kidney, lungs, nasal cavity, prostate gland, gastrointestinal tract, and bladder cancers) and cardiovascular dysfunction have also been found. In a limited area on the southwest coast of Taiwan, blackfoot disease (BFD) as shown in Figures 1 and 2, a peripheral vascular disease, has been attributed to arsenic ingestion. BFD refers to the progressive discoloration of the extremities which may, in severe cases, eventually necessitate amputation (Chen et al., 1994).



Figure 1 The two kids had the blackfoot disease. They were nine and six years old, respectively (Wang and Ren, 2004).



Figure 2 A married woman, 24 years old, had the blackfoot disease (Wang and Ren, 2004).

The World Health Organization has estimated that a lifetime exposure to inorganic arsenic in drinking water at a concentration of $200 \mu\text{g/L}$ is associated with a 5% risk of skin cancer. Arsenic is classified by the USEPA as a class A (i.e., known) human carcinogen, thus the anticipated maximum contaminant level goal (MCLG) is zero; which analytically means below the detection limit of the most sensitive available analytical techniques. Recent studies have indicated, however,

that the dose-response relationship between cancer and arsenic exposure may not be linear and that a threshold, related to arsenic detoxification in the body, may exist (Petito and Beck, 1990; Thompson, 1993). USEPA Risk Assessment Council (RAC) has also completed its health assessment for non-cancer effects and a reference dose of $0.3 \mu\text{g}/\text{kg}/\text{d}$ has been adopted (range of 0.1 to $0.8 \mu\text{g}/\text{kg}/\text{day}$). The drinking water equivalent level (DWEL) is calculated to be $10.5 \mu\text{g}/\text{L}$ (assuming 70kg body weight and $2\text{-L}/\text{d}$ consumption). Use of a default value of 20 percent for the relative source contribution (RSC) for arsenic was initially proposed; the anticipated maximum contaminant level goal (MCLG) for no cancer effects would be about $2 \mu\text{g}/\text{L}$, a 25-fold decrease from the MCL value of $50 \mu\text{g}/\text{L}$ (Pontius, 1993).

For most of the population, the main route of exposure to inorganic arsenic is through ingestion of drinking water. The international drinking water standards are in the range of 10 (i.e., World Health Organization) to $50 \mu\text{g}/\text{L}$ arsenic. Arsenic concentrations in bottled drinking water are also regulated (CFR, 1992). Unquestionably, adoption of more stringent standards would necessitate treatment of many drinking water supplies for arsenic removal. Thus, compliance cost and treatment technologies are major concerns of water agencies.

Arsenic ranks 20th in elemental abundance in the earth's crust with an average crustal abundance of 1.8 ppm (National Research Council, 1997) and occurs as both stable oxide and sulfide minerals (Table 1). The elevated concentrations of arsenic in groundwaters and seepage-fed surface waters may be derived naturally from the leaching of arsenic-rich soil and aquifer minerals. Human activities can also contribute to high concentrations and enhance mobilization of arsenic in potential source waters for drinking water supplies. Increased arsenic mobilization can be caused by agricultural practices such as exhaustive irrigation and use of phosphate-containing fertilizers (Peryea, 1991; Davenport and Peryea, 1991). Soils have been polluted by arsenic through use of arsenical pesticides (Table 1), leaching of mine tailings, or deposition of arsenic originally released into the atmosphere by metal smelting and coal combustion (Ferguson and Gavis, 1972; Chilvers and Peterson, 1987; Bodek et al., 1988; Mok and Wai, 1989; Seyler and Martin, 1989; Davenport and Peryea, 1991). Arsenic may also be introduced into ground and surface waters by discharge of contaminated wastewater effluents or disposal of chemical wastes (Jekel, 1994).

There are many cases of arsenic occurring in high concentrations in ground water in certain countries, such as Taiwan, Germany, Chile, India and the United States. Elevated arsenic concentrations (50-48,000 $\mu\text{g/L}$) have been found in ground water in the western United States and are associated with mining areas, geothermal areas, and basin-fill deposits (Welch et al., 1988). High arsenic concentrations (at the ppm level) have been found in tap water in Taiwan (Shen, 1973; Hsia et al., 1992).

Table 1 Chemical forms of arsenic in natural minerals, arsenic pesticides, natural biogenic As compounds and major anthropogenically -produced As forms.

Name	Chemical formula
<u>Natural minerals</u>	
scorodite	$\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$
arsenopyrite	FeAsS
realgar	AsS
orpiment	As_2S_3
arsenolite	As_4O_6
<u>Natural biogenic As compounds</u>	
arsenobetaine	$(\text{CH}_3)_3\text{As} + \text{CH}_2\text{COOH}$
arsenochlorine	$(\text{CH}_3)_3\text{As} + \text{CH}_2\text{OH}$
<u>Arsenic pesticides</u>	
herbicides and pesticides	$\text{CHAsO}_3\text{HNa}, \text{CHAsO}_2\text{Na}_2, (\text{CH}_3)_2\text{AsO}_2\text{Na}$
weed killer	H_3AsO_4
insecticides	$\text{Pb}_3(\text{AsO}_4)_2, \text{Ca}_3(\text{AsO}_4)_2$
<u>Anthropogenically-produced As forms</u>	
Gold mining area-may be scorodite or arseniferous pyrite	
Gold ore-orpiment (As_2S_3), realgar (AsS), arsenopyrite (FeAsS)	
Smelting and Mining operations-emission of arsenical particulates (typically As_2O_3)	
References: Anderson and Bruland, 1991; Welch et al., 1988; Woolson, 1977; Cullen and Reimer, 1989.	

The occurrence of arsenic in source waters for municipal drinking water supplies combined with more stringent regulation may necessitate widespread treatment for arsenic removal. The standard of the current maximum contaminant level (MCL), 10 $\mu\text{g/L}$, for arsenic is being regulated in law by the USEPA under court-ordered deadline. Enhanced

coagulation is one of the applicable technologies for arsenic removal. Although enhanced coagulation is a relatively economical process, high compliance costs have been estimated even for a new MCL range of 2-20 μ g/L. Costs estimated by the USEPA are approximately \$140 million for compliance with a MCL of 20 μ g/L to \$6.2 billion in capital investment and \$1 billion annually to meet the lower standard of 2 μ g/L (Reid, 1994; Pontius and Roberson, 1994). Assessment of the efficiency of arsenic removal by various technologies, including enhanced coagulation, is crucial both to establish a new arsenic MCL and to satisfy future standards.

1.1 Mechanisms of contaminant removal by coagulants

There are a variety of mechanisms involved in contaminant removal during coagulation treatment. With hydrolyzing metal salts as coagulants, the aqueous chemistry of the metals (i.e., in hydrolysis, adsorption, polymerization, and precipitation reactions) plays a critical role in contaminant removal.

Coagulation diagrams are commonly used to predict (or rationalize) optimal coagulant dosages and pH ranges for coagulation. These diagrams bear a marked similarity to solubility diagrams for amorphous aluminum oxides. The applicability of coagulation diagrams to a specific source water is influenced by source water characteristics such as temperature, suspended solids concentration (turbidity), and chemical composition.

The dominant mechanism(s) for contaminant removal depend on the type of contaminant (e.g., particles, inorganic contaminants, DOM), the type and concentration of the coagulant, and other factors such as pH, ionic strength, and major ion composition of the source water. A mechanistic understanding of contaminant removal is necessary to explain coagulation data and to determine the appropriate dosage and type of coagulation and treatment conditions for a particular source water. Contaminant removal can be achieved by several important mechanisms: (1) double-layer compression, (2) adsorption and charge neutralization, (3) enmeshment in precipitates, (4) interparticle bridging, (5) solid-solution formation, (6) surface precipitation, (7) ligand exchange-surface complexation, and (8) hydrogen bonding.

1.2 Chemistry of hydrolyzing metal salts

The most widely used coagulants for water treatment in the United States are aluminum salts. The mechanism of contaminant removal depends on the speciation of aluminum and the extent of supersaturation with respect to the hydroxide solid during water treatment. Figure 3 illustrates the solubility diagram for amorphous aluminum hydroxide. The concentration of monomeric and polymeric species and the region of solid precipitation are shown as a function of pH. Equilibrium formation constants used for this solubility diagram are given in Table 2. The amorphous aluminum hydroxide is observed to be least soluble near neutral pH (Stumm and O'Melia, 1968; Amirtharajah and O'Melia, 1990).

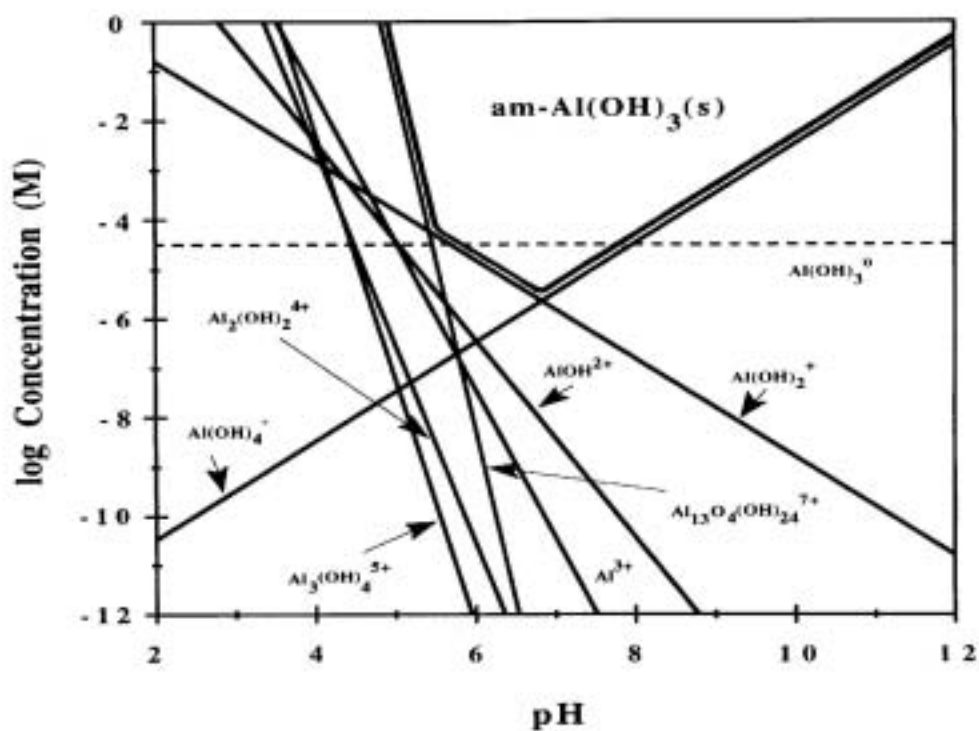


Figure 3 Stability diagram for amorphous aluminum hydroxide

Table 2 Equilibrium constants for reactions of aluminum (25°C)

The concentrations of all aluminum species is written as an explicit function of $[H^+]$

$$[Al_x(OH)_y^{(3x-y)+}] = \frac{K_{xy}[Al^{3+}]^x[H^+]^{-y}}{[H^+]^{3x-y}}$$

Aluminum species	Formation constants Log $K_{xy}(I=0.001M)$	x	y	Reference
$AlOH^{2+}$	-5	1	1	(1)
$Al(OH)_2^+$	-9.039	1	2	(2)
$Al(OH)_3^0$	-15.09	1	3	(2)
$Al(OH)_4^-$	-23.06	1	4	(2)
$Al_2(OH)_2^{4+}$	-7.7	2	2	(2)
$Al_3(OH)_4^{5+}$	-13.99	3	4	(1)
$Al_{13}(OH)_{24}^{7+}$	-103.9	13	NA	(1)
$Al(OH)_3(s)$	-10.59	1	3	(3)

References: (1) Ohman and Forsling, 1981

(2) Baes and Mesmer, 1976

(3) Amirtharajah and O'Melia, 1990

NA: Not Applicable

2. Materials and methods

2.1 Materials-Standards and Reagents

All chemicals were reagent grade (or better) and were used without additional purification. All solutions were prepared with de-ionized water (Barnstead Nanopure II, Dubuque, IA), pre-treated by reverse osmosis. All glassware was cleaned by soaking in 6% HCl and rinsed 4 times with de-ionized water. Arsenic (III) primary stock solutions (1g As/L in 0.18 M HCl) were prepared from solid arsenic trioxide, As_2O_3 (Aldrich, A.C.S. primary standard). The arsenic (V) primary stock solutions (1g As/L in 0.18 M HCl) were prepared from sodium salt heptahydrate, $Na_2HAsO_4 \cdot 7H_2O$ (Sigma). All dilution and stock solutions were made with 0.18 M (1.5% v/v) Trace Metal grade HCl (Fisher) and de-ionized (DI) water. Hydride generation was accomplished using analytical grade 4% (w/v) sodium borohydride pellets (Alfa Products) dissolved in 1% (w/v) sodium hydroxide (Fisher). Ultra pure 0.14 M potassium iodide (Alfa) was used as a pre-reductant for As (V) analysis. Atomic grade acetylene (Liquid air) and 99.999% nitrogen (Liquid carbonic) were used as the AAS flame fuel for the atomic absorption spectrometer furnace and MHS-10 purge gas for the hydride generation unit, respectively.

A stock solution of alum (aluminum sulfate) was prepared by dissolving analytical reagent-grade $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ (Fisher Scientific, Pittsburgh, PA) in de-ionized water to a concentration of 0.3 M (199.8 g $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$, 0.6 M as aluminum). In all coagulation experiments, a fresh stock solution was prepared and was stored in a refrigerator at 4°C until use. Alum was added directly from the stock without intermediate dilution. Irreproducible results have been reported with use of dilute and aged stock solutions (Ching, 1994). Note that 1 μm of aluminum is equivalent to 0.33 mg/L of alum, $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$.

Background electrolyte solutions (0.01 M NaNO_3 , 0.001 M NaHCO_3) were prepared from the reagent grade salts NaNO_3 and NaHCO_3 (Fisher) and a primary calcium stock solution (0.1M Ca^{2+} or 4.0 g Ca/L) was from $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (Fisher) and a primary calcium stock solution (0.01M SO_4^{2-} or 0.96 g SO_4/L) was from $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ (Fisher). A primary (0.01M PO_4^{3-} or 0.95 g PO_4/L) and secondary phosphate stock solutions (10^{-4} M PO_4^{3-} or 9.5 mg PO_4/L) were prepared from $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$ (Fisher). Kaolin stock solution (500 mg/L) was prepared in de-ionized water using USP grade kaolin (Fisher) without further purification. Kaolin stock solutions were sonicated for 30 min (BRANSON B-220, Smithkline) before use.

2.2 Conditions and apparatus for coagulation experiments

Experiments were conducted with source waters or with background electrolyte 0.01 M NaNO_3 and 0.001 M NaHCO_3 (pH before adjustment 8.3 ± 0.1). The experiments were performed at room temperature (18 to 23 °C). Test solutions were contained in 1 L cylindrical Pyrex beakers and mixed using a high speed, six paddle stirrers (Fisher). For coagulation experiments, the pH of 1 L of source waters or amended and/or un-amended background electrolyte was pre-adjusted to the desired value (± 0.1 pH unit) by addition of HCl or NaOH. Solutions were then spiked with As (III) or (V) to the desired concentration. All adsorbates were added prior to coagulant addition. Pre-determined concentrations of base (to maintain the target pH) were added simultaneously with aliquots of 0.3 M alum coagulant stock solutions under rapid-mix conditions (mixing conditions: 1 min rapid-mix at 100 rpm ($G=160\text{s}^{-1}$), 30 min slow mix at 45 rpm ($G=48\text{s}^{-1}$), 15 min settling). After the settling period, ca.100mLs of solution was collected using a 20 mL disposable syringe from 2 cm below the liquid surface and filtered through a 0.1 μm cellulose nitrate membrane filter (Sartorius Edgewood, NY) using a vacuum filter flask assembly. The filter flask assembly was rinsed 3 times with DI water between samples; filters were not re-used. In some experiments, disposable syringe filter assemblies (Anotop, Maidstone, England) were employed; syringe filter assemblies were not re-used.

The pH was recorded after the settling period. Filtrate from coagulation

experiments was transferred to 60 mL, acid-washed HDPE bottles and acidified by additions of 1.0 mL concentrations in this study. Losses of As (III) and (V) to coagulation and filtration apparatus were assessed in blank loss experiments. Except for the absence of the coagulant, the same experimental conditions were used in the blank loss experiments as in the coagulation experiments

3. Results and Discussions

3.1 Aluminum solubility

The solubility of amorphous aluminum hydroxide must be exceeded to provide a substrate for arsenic adsorption though this is not strictly necessary for turbidity removal. To test the completeness of precipitation of added aluminum, overlying solutions of background electrolyte from alum coagulation experiments conducted with 20 and 80 mg alum/L (corresponding to 60 and 240 μM total aluminum) were collected after settling and filtered through 0.1 μm membrane filters. The filtered solutions were analyzed for total aluminum. Figure 4 shows the percent aluminum retained by 0.1 μm filter with pH after coagulation with 20 and 80 mg alum/L and filtration. It was observed that close to quantitative removal of aluminum can be achieved only in the pH 6-7 range; at higher or lower pH values, the percent aluminum removed decreased more rapidly for the lower alum dose. Some discrepancies were observed between initial aluminum concentrations and measured aluminum concentrations at both extreme pH values (4 and 9); such discrepancies are most probably due to experimental errors in sample dilution.

The extent to which added aluminum is precipitated from solution is strongly dependent on pH and coagulant dose. The observed trend in retention of aluminum by filters was consistent with predictions of precipitation of amorphous aluminum hydroxide obtained with MINEQL⁺ (Hering and Elimelech, 1995). Previous studies of aluminum solubility have demonstrated substantial precipitation of amorphous aluminum hydroxide over a pH range of 5.5 to 7.5 with 200 mg alum/L. The pH of most rapid precipitation (pH =7.5) was found to correspond to the pH of most efficient coagulation of mineral suspensions by alum (Packham, 1965). In this study, the principal concern is that adsorption of arsenic onto the surface of freshly-precipitated amorphous aluminum hydroxide cannot occur unless the

solubility of the solid is exceeded. Thus, the range of coagulant dose and pH for arsenic removal are limited by solubility considerations.

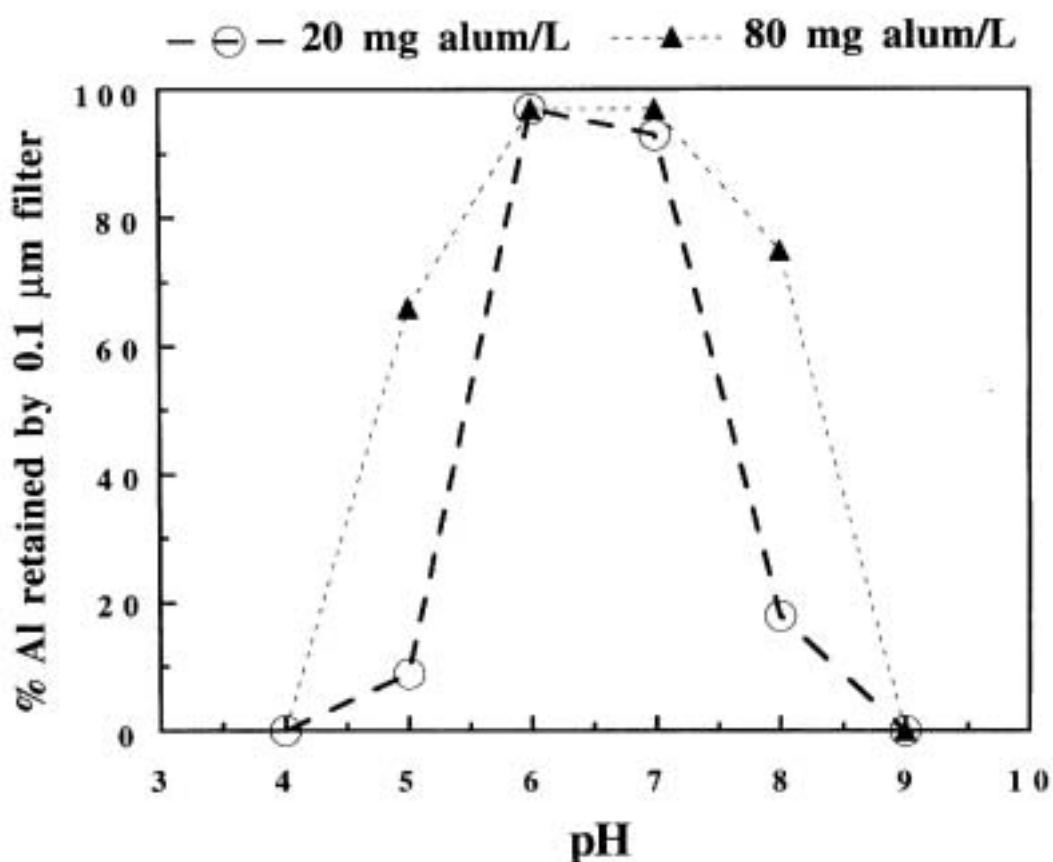


Figure 4 Fraction of added aluminum precipitated as amorphous $\text{Al}(\text{OH})_{3(s)}$ in pH-adjusted background electrolyte (0.01 M NaNO_3 , 0.001 M NaHCO_3) as indicated by retention of aluminum by membrane filters after coagulation with 20 and 80 mg alum/L

3.2 Effect of coagulant dose and arsenic oxidation state

The influence of varying the coagulant dose on As removal from background electrolyte pH-adjusted to 6.0 and 7.0 by coagulation with alum is illustrated in Figure 5 for both As(III) and (V) with initial As concentrations of 9 and 20 $\mu\text{g}/\text{L}$, respectively. Complete removal of As (V) was observed for coagulant doses above 5 mg alum/L at pH 6. Even under conditions where the added aluminum is incompletely precipitated as amorphous aluminum hydroxide (ca. 20 mg alum/L and pH 8), the observed removal was 87%; at lower coagulant dose (5 and 10 mg alum/L), above 30% removal of As (V) was still obtained. During

the course of study, it was established that the dosage of alum coagulant needed to achieve high As (V) removal was often far below that required to produce a floc that settled well.

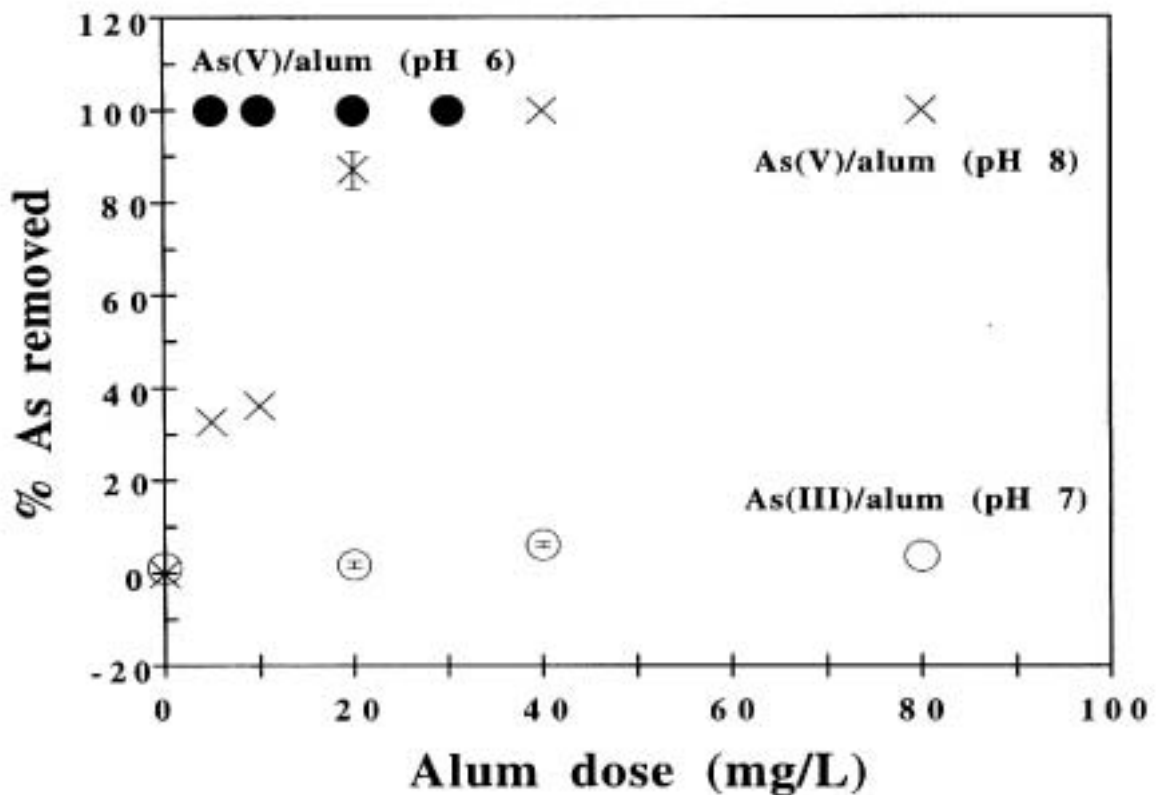


Figure 5 Removal of arsenic from pH-adjusted background electrolyte (0.01 M NaNO₃, 0.001 M NaHCO₃) by coagulation with alum is shown for As (III) and As (V) as a function of alum dose (mg/L).

In contrast, extremely poor removal of As(III) was observed with the range of coagulant doses studies, up to ca. 80 mg alum/L at pH 7.0 (Figure 5).

Negligible As (III) removal was observed even with considerably higher alum doses of up to 300 mg/L. Interpretation of observations for negligible As(III) removal over the pH range 4 to 8.5 at various alum doses (5 to 300 mg alum/L) may be somewhat complicated by solubility considerations. Nor was As (III) adsorption observed in the adsorption experiments with hydrous aluminum oxide in the absence of sulfate (Hering and Elimelech, 1995). Thus, initial As oxidation state has even more effect on As removal by alum than previously observed with ferric chloride.

3.3 Effect of initial As (V) concentration

Removal of As (V) from pH-adjusted background electrolyte (0.01 M NaNO₃ and 0.001 M NaHCO₃) by alum was examined at pH 6.0 with a coagulant dose of 20 mg alum/L and at pH 8.5 with 30 mg alum/L for initial As (V) concentrations ranging from 2 to 100 µg/L (Figure 6). Complete removal of As (V) was observed at pH 6.0 (20 mg alum/L). At pH 8.5, As (V) removal was poor (between 40 and 60%) over most of the concentration range studied. Significantly better removal (87%) was observed at the lowest initial As (V) concentration tested. Interpretation of observations at pH 8.5 is, however, complicated by the incomplete precipitation of added aluminum as amorphous aluminum hydroxide at this pH.

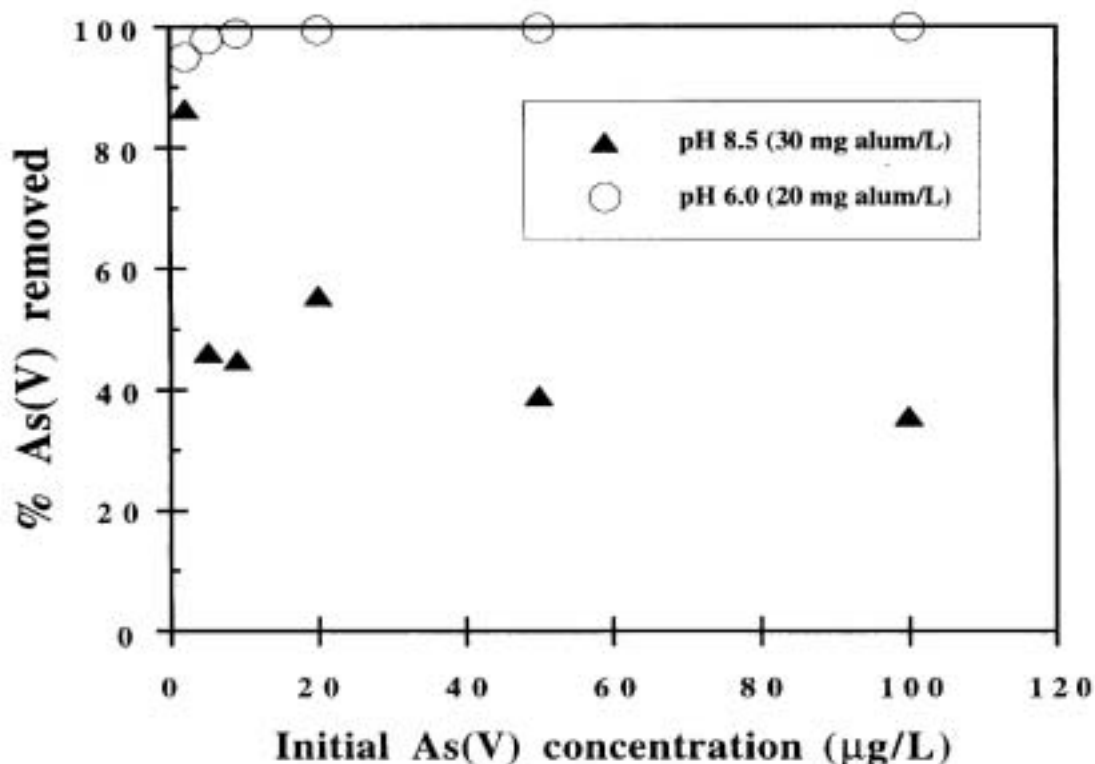


Figure 6 Effect of initial As (V) concentration (2-100 µg/L) on As (V) removal from background electrolyte at pH values of 6.0 and 8.5 and coagulant doses of 20 and 30 mg alum/L.

As discussed before, the lack of dependence of percent As (V) removal on initial As (V) concentration in the lower concentration range is consistent with adsorption of As onto the surface of freshly-precipitated amorphous aluminum oxide at low surface coverage.

3.4 Effect of pH on As (V) removal

As described in Section 3.1, the greater solubility of amorphous aluminum hydroxide restricts the pH range for this system. The variations in As (V) removal from background electrolyte (0.01 M NaNO₃ and 0.001 M NaHCO₃) by coagulation with alum as a function of pH are illustrated in Figure 7. As shown in Figure 7, which total aluminum concentration of 60 μM Al (20 mg alum/L) and 120 μM Al (40 mg alum/L), the As(V) removal is close to complete over the pH range 5 to 8 but decreases markedly at higher pH values with the lower alum dose (ca. 20 mg alum/L). As mentioned in Section 3.3, however, decreased As removal under these conditions may be due partly to incomplete precipitation.

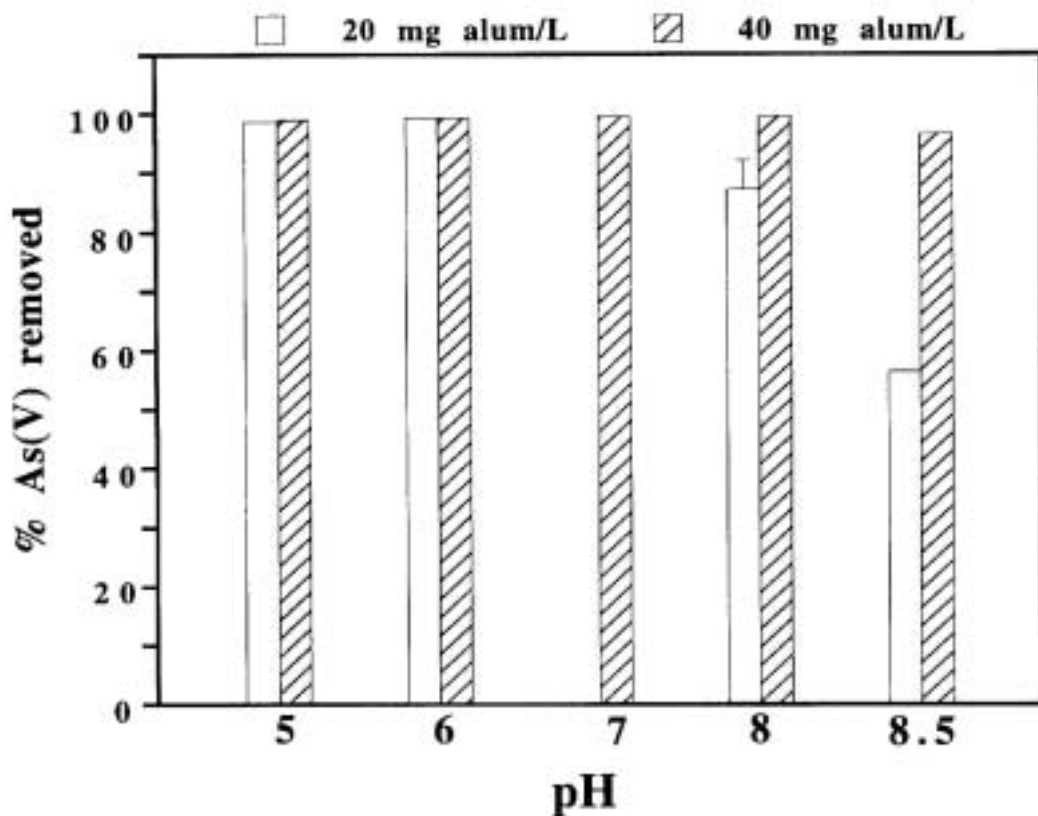


Figure 7 Effect of pH on As (V) removal from background electrolyte with initial As (V) concentration of 20 μg/L and alum doses 20 mg/L and 40 mg/L.

4. Conclusions

The removal of arsenic by amorphous aluminum hydroxide precipitated *in situ* has been studied as a function of various parameters. Adsorption of As (V) onto the surfaces of freshly-precipitated amorphous aluminum oxides has been confirmed as the dominant mechanism that governs removal of As (V) in the coagulation processes.

This study supports the conclusion that residual As (V) concentrations in treated water below 2 µg/L can be obtained by coagulation with alum (at lower pH ca.6.0 or 7.0) for ambient influent As (V) concentrations in the range 2 to 100 µg/L. On a molar basis, comparable As (V) removal was observed by coagulation with alum.

Water treatment plants treating source waters containing As (III), such as reducing groundwaters (Ferguson and Gavis, 1972; Sorg and Logsdon, 1978), should pre-oxidize As (III) to As (V) for most efficient removal. However, alum cannot be used for As (III) removal.

As (V) removal by alum as a function of pH typically decreases sharply over a narrow range of 1 to 2 pH units. As (V) removal by alum (20 mg/L) decreased from 87% to 56% from pH values 8 to 8.5.

One additional limitation of the study described in this chapter is solubility of amorphous aluminum hydroxide at both extreme pH values and low alum doses. In practice, many researchers have reported that better As (V) removal was achieved in bench-scale tests than at the pilot scale with alum treatment, whereas similar As (V) removal was obtained at the bench scale, pilot scale and full scale for iron coagulation (Sorg, 1993; Scott et al., 1995).

The application of this study is limited to the simpler systems studied. The understanding of interactions of inorganic solutes and natural organic matter with coagulants is clearly needed to characterize fully the potential impact of water composition on arsenic removal by coagulation processes.

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