



Arsenic removal from groundwater by acclimated sludge under autohydrogenotrophic conditions

Siqing Xia*, Shuang Shen, Xiaoyin Xu, Jun Liang, Lijie Zhou

State Key Laboratory of Pollution Control and Resource Reuse, College of Environmental Science and Engineering, Tongji University, Shanghai 200092, China

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ABSTRACT

Arsenic in the environment is attracting increasing attention due to its chronic health effects. Although arsenite (As(III)) is generally more mobile and more toxic than arsenate (As(V)), reducing As(V) to As(III) may still be a means for decontamination, because As(III) can be removed from solution by precipitation with sulfide or by adsorption or complexation with other metal sulfides. The performance of As(V) bio-reduction under autohydrogenotrophic conditions was investigated with batch experiments. The results showed that As(V) reduction was a biochemical process while both acclimated sludge and hydrogen were essential. Most of the reduced arsenic remained in a soluble form, although 20% was removed with no addition of sulfate, while 82% was removed when sulfate was reduced to sulfide. The results demonstrated that the reduced arsenic was re-sequestered in the precipitates, probably as arsenic sulfides. Kinetic analysis showed that pseudo first-order kinetics described the bio-reduction process better than pseudo second-order. In particular, the influences of pH and temperature on As(V) reduction by acclimated sludge under autohydrogenotrophic conditions and total soluble As removal were examined. The reduction process was highly sensitive to both pH and temperature, with the optimum ranges of pH 6.5-7.0 and 30-40°C respectively. Furthermore, Arrhenius modeling results for the temperature effect indicated that the As(V) reduction trend was systematic. Total soluble As removal was consistent with the trend of As(V) reduction.

Introduction

Arsenic, a known carcinogen in humans, is often found in contaminated groundwater as a result of weathering of rocks, industrial waste discharges, agricultural use of arsenical herbicides and pesticides, etc. (Bhumbla and Keefer, 1994). Exposure to arsenic-contaminated drinking water has caused serious health problems in various countries, including Japan (ATSDR, 2002), Bangladesh (Wasserman et al., 2004), Shanxi Province, China (Wang et al., 2007) and Bengal (Guha et al., 2004). Because of the health effects of arsenic, most countries in the world have adopted the guideline of 10 μ g/L for public water systems (WHO, 2001). Arsenic is a metalloid (or inorganic semi-conductor) that can occur with oxidation states of -3, 0, +3, and +5. The As(V) and As(III) forms are the most common in natural waters (Cullen and Reimer, 1989; Jekel, 1994). The ratio of the concentrations of As(V) and As(III) is controlled primarily by redox potential and pH (Tallman and Shaikh, 1980; Vance and Jacobs, 1995). At pH values of most natural waters and under reducing conditions, As(III) is present as non-ionic arsenious acid (H₃AsO₃). On the other hand, As(V) dominates under oxidizing conditions and is mainly present in the oxyanionic forms (H₂AsO₄⁻, HAsO₄²⁻) (Cullen and Reimer, 1989b; Smedley et al., 1996; Yan et al., 2000).

Physical and chemical methods are the main treatment processes for arsenic-contaminated water, including anion exchange (Korngold et al., 2001; Vagliasindi and Benjamin, 1998), reverse osmosis (US EPA, 1999), chemical

^{*} Corresponding author. E-mail: siqingxia@gmail.com

precipitation (Borho and Wilderer, 1996; Wickramasinghe, 2004; Tipping, 1981; Meng et al., 2002) and adsorption (Soner et al., 2002; Guha and Chaudhuri, 1990). In addition to physical and chemical methods, the activated sludge process and use of bacteria algal symbionts were two typical biosorption methods for arsenic removal (Goldstone et al., 1990; Jahan et al., 2006). In terms of microbial processes, Luo et al. (2008) reported one of sulfate-reducing bacteria that can reduce sulfate to sulfide as well as As(V) to As(III), so that arsenic can be effectively removed as Assulfide insoluble precipitate. Lim et al. (2008) studied the iron-reducing bacterium Shewanella sp., which can reduce As(V) to As(III) with an organic electron donor. The ability of sulfate-reducing bacteria to reduce arsenic as well as other metals and metalloids has been well documented (Jong and Parry, 2003; Simonton et al., 2000).

The bio-reduction of As(V) can occur with a number of different electron donors. Many researchers have used organic electron donors, such as acetate, lactate, pyruvate, butyrate, and ethanol (Luo et al., 2008; Lim et al., 2008; Jong and Parry, 2003; Oremland et al., 1994; Dowdle et al., 1996; Macy et al., 1996; Newman et al., 1997a, 1997b; Blum et al., 1998; Harrington et al., 1998; Stolz and Oremland, 1999). Nevertheless, using an organic electron donor not only significantly increases excess sludge production, but also involves handling of chemicals that are expensive (Xia et al., 2009). Compared with organic electron donors, H₂ shows advantages as an ideal electron donor, including nontoxicity and lower production of excess biomass (Rittmann et al., 2004). Many oxidized contaminants can be reduced to less toxic or less mobile species by acclimated microorganisms under autohydrogenotrophic conditions (Nerenberg, 2005). The classical example is nitrate, which can be reduced to nitrite and nitrogen gas stepwise (Nerenberg and Rittman, 2004). Recently, more and more oxidized contaminants were shown to be reduced. For instance, bromate (BrO_2^{-}) can be reduced to Br⁻ ion (Downing and Nerenberg, 2007); selenate (SeO_4^{2-}) can be reduced to less mobile elemental selenium (Se°) (Chung et al., 2006a); and chromium can be reduced from hexavalent chromate (CrO_4^{2-}) to less toxic Cr^{3+} , which precipitates as $Cr(OH)_3$ and is removed from solution (Chunget al., 2006b).

In this article, we mainly investigated the feasibility of arsenate bio-reduction by acclimated sludge under autohydrogenotrophic conditions, as well as its kinetics. The effects of some key factors like pH and temperature on the bio-reduction process were explored. The precipitate was analyzed to determine whether the reduced arsenate was precipitated.

1 Materials and methods

1.1 Experimental setup

Plastic physiologic saline bottles with the effective volume of 250 mL were used as the reactors. The cap was butyl synthetic rubber, with air tightness, and the solution volume was 200 mL, including feed medium and inoculation sludge. Stock solution was added into the reactor with a sterile syringe. Residual air in the reactor was expelled through a syringe needle, and hydrogen was injected through another needle linked to a hydrogen tank. The reactor was covered with aluminum foil and incubated on a shaking table at 160 r/min. Each time after sampling, any residual air in the bottles was expelled, and new hydrogen gas was injected to ensure adequate supplies of hydrogen through the reaction process.

1.2 Feed medium and stock solution

The composition of the feed medium was as follows: (in g/L) NaNO₃ 0.060, NaHCO₃ 0.016, Na₂HPO₄·12H₂O 4.441, KH₂PO₄ 1.035, FeSO₄·7H₂O 0.001, CaCl₂·2H₂O 0.001; and (in mg/L) ZnSO₄·7H₂O 0.013, H₃BO₃ 0.038, CuCl₂·2H₂O 0.001, Na₂MoO₄·2H₂O 0.004, MnCl₂·4H₂O 0.004, CoCl₂·6H₂O 0.025, NiCl₂·6H₂O 0.001. Nitrate, as the most common pollutant in groundwater, was added to cultivate the microorganisms. NaHCO3 was added as an inorganic carbon source for the microorganisms and phosphate buffer $(KH_2PO_4 + Na_2HPO_4)$ was used to prevent any sharp pH rise, since denitrification and the reduction of sulfate as well as arsenate may cause a significant rise of pH value. All feed media were purged with N₂ gas to eliminate dissolved O_2 . The pentavalent arsenic stock solution contained deionized water with 1000 mg/L of As(V) (from Na₃AsO₄·12H₂O).

1.3 Inoculum, starting up, and feasibility research

Anerobic sludge, as inoculation sludge, was obtained from the anoxic pond of Quyang Sewage Treatment Plant (Shanghai, China). The sludge concentration was about 3000 mg/L and the ratio of volatile suspended solids (VSS) to suspended solids (SS) was 65%-70%. Inoculation sludge (20 mL) was first rinsed three times with a sodium phosphate buffer ($KH_2PO_4 + Na_2HPO_4 + KCl + NaCl$), then added into the reactor, which contained the feed media and 10 mg N/L nitrate to accumulate microorganisms. Once nitrate and nitrite were completely removed, the denitrification process, which was maintained for 24 hr, was finished, and during the whole acclimation process the pH did not change sharply due to the presence of phosphate buffer (< 0.1 pH unit); then pentavalent arsenic stock solution was added into the reactors. To investigate the potential for bio-reducing As(V), a test group and four control groups were examined. The test group was investigated with three replications, with starting As(V) concentrations 2 mg/L and sulfate 30 mg/L, to ensure the reliability of results. The four control groups were investigated without Download English Version:

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