Chemosphere 194 (2018) 49–56

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Characterising microbial reduction of arsenate sorbed to ferrihydrite and its concurrence with iron reduction

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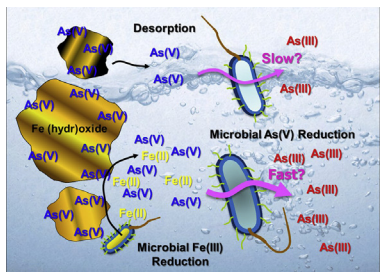
HIGHLIGHTS

- The dissolved As(V) availability governed the reduction kinetics of sorbed As(V).
- High lactate concentrations may slow down the As(V) reduction rate.
- As(V) and ferrihydrite reduction by *S. putrefaciens* occurred simultaneously.
- Formation of ferrous arsenate may retard microbial arsenate reduction.
- Ferrihydrite reductive dissolution quickened As(V) reduction at high As:Fe ratios.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 September 2017
Received in revised form
6 November 2017
Accepted 20 November 2017
Available online 21 November 2017

Keywords:
Arsenate reduction
Kinetics
Ferrihydrite
Shewanella

ABSTRACT

A series of model anoxic incubations were performed to understand the concurrence between arsenate and ferrihydrite reduction by *Shewanella putrefaciens* strain CN-32 at different concentrations of arsenate, ferrihydrite and lactate, and with given ΔG_{rxn} for arsenate and ferrihydrite reduction in non-growth conditions. The reduction kinetics of arsenate sorbed to ferrihydrite is predominately controlled by the availability of dissolved arsenate, which is measured by the integral of dissolved arsenate concentrations against incubation time and shown to correlate with the first order rate constants. High lactate concentrations slightly slowed down the rate of arsenate reduction due to the competition with arsenate for microbial contact. Under all experimental conditions, simultaneous arsenate and ferrihydrite reduction occurred following addition of *S. putrefaciens* inoculums and suggested no apparent competition between these two enzymatic reductions. Ferrous ions released from iron reduction might retard microbial arsenate reduction at high arsenate and ferrihydrite concentrations due to formation of ferrous arsenate. At high arsenate to ferrihydrite ratios, reductive dissolution of ferrihydrite shifted arsenate from sorption to dissolution and hence accelerated arsenate reduction. The interaction between microbial arsenate and ferrihydrite reduction did not correlate with ΔG_{rxn}, but instead was governed by other factors such as geochemical and microbial parameters.

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1. Introduction

Elevated arsenic (As) concentrations in natural waters render a major problem with water quality and health for millions people worldwide, e.g. in Bangladesh, Vietnam and United States (Garelick et al., 2005). Due to high toxicity of As, the drinking water limit of As concentration is very low in many countries (50 or 10 μg L^{-1}) (EU, 1998; WHO, 2001); however Cantor (1996) recommended even lower concentrations to be tolerable. Naturally high

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https://doi.org/10.1016/j.chemosphere.2017.11.109
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concentrations of As in surface and ground waters may be due to geothermal sources, the dissolution of As-bearing minerals in soils and sediments or large anthropogenic inputs from emissions of mining and smelting activities (Smedley and Kinniburgh, 2002; Niazi et al., 2018). In the As calamity in Bangladesh, West Bengal and Southeast Asia, young sediments are the source of high As levels in shallow ground waters, where As release is mostly attributed to microbial reductive dissolution of Fe(III) phases that retain As and the simultaneous reduction of arsenate (As(V)) to arsenite (As(III)) (Zobrist et al., 2000; Islam et al., 2004, 2005; Weber et al., 2010; Huang et al., 2011b; Huang, 2014).

A number of field studies indicated a strong correlation between As mobilisation and reductive dissolution of Fe(III) (hydr)oxides in reducing soils and sediments (Islam et al., 2004; Weber et al., 2010). While Fe(III) (hydr)oxides is the primary sorbent for As, As can be released into the solution when the Fe(III) (hydr)oxides are reduced and then dissolve (Cummings et al., 1999; Burnol et al., 2007; Karimian et al., 2017; LeMonte et al., 2017). On the other hand, model experiments show that microbial reduction of Fe(III) (hydr) oxides may lead to formation of Fe(II)-bearing phases that are able to capture As species (Islam et al., 2005; Coker et al., 2006), thus repartitioning Fe and As from solution to solid phase. In this case, microbial As(V) reduction becomes the major driving force for As mobilisation. Therefore, under reducing condition, the interplay between microbial As(V) and Fe(III) (hydr)oxide reduction determines the mobilisation of solid associated As and As ecotoxicity. Based on reaction free energy, microbial As(V) reduction has to compete with other reducing reactions such as nitrate and sulphate reduction (Dowdle et al., 1996; Newman et al., 1997). Thermodynamically, reduction of crystalline Fe (hydr)oxides such as goethite and hematite is less favourable than reduction of dihydrogen As(V) (Kocar and Fendorf, 2009). In comparison, reduction of amorphous Fe (hydr)oxides such as ferrihydrite reduction yields similar Gibbs free energy as reduction of As(V) (Campbell et al., 2006). Since the periodic redox fluctuations of As in soils is often associated with amorphous Fe (hydr)oxides (Huang and Kretzschmar, 2010), it is important to understand the priority of As(V) and ferrihydrite reduction and the interplay between these two reactions.

To date, microbial reduction of different Fe(III) (hydr)oxides have been well characterised e.g. (Liu et al., 2001; Roden, 2003). Many groups have reported on microbial reduction of As(V) in solution (Newman et al., 1997; Zobrist et al., 2000; Yamamura et al., 2003; Huang et al., 2010). The primary focus in the literature concerning Fe(III) and As(V) reduction was to understand the mobilisation of As(V) in microbial reducing processes, however mostly at relatively high As concentrations (mM level) (Zobrist et al., 2000; Islam et al., 2005; Coker et al., 2006; Burnol et al., 2007). In comparison, microbial respiratory reduction of solid associated As(V) and the potential influence of the simultaneous Fe(III) reduction under different environmental conditions is less understood due to the difficulty to speciate solid associated As. Moreover, it is not clear about the order of initialisation of As(V) and ferrihydrite reduction by microbes in relation to Gibbs free energy of reaction. An analytical method with sufficient sensitivity and reproducibility is critical for monitoring the kinetics of As(V) reduction due to low As(III) concentration and subtle decrease of As(V) concentrations in the initial state. In light of the new extraction method developed by Huang and Kretzschmar (2010), the reduction kinetics of solid associated As(V) can be investigated with not only higher sensitivity but also better time resolution as compared to synchrotron based X-ray absorption spectroscopy (Weber et al., 2010). Therefore, this study was aimed to characterise microbial reduction kinetics of As(V) sorbed to ferrihydrite at different As(V) to ferrihydrite ratios with the range of As(V) concentration representing normal to high As contaminated environments (Smedley and Kinniburgh, 2002; Islam et al., 2004; Huang and Matzner, 2006, 2007; Huang and Kretzschmar, 2010; Wang et al., 2012), and the relationship of ferrihydrite and As(V) reduction simultaneously driven bacterial respiration mechanism. Specifically, this investigation will (1) identify the parameters governing microbial reduction kinetics of As(V) sorbed to ferrihydrite, (2) determine how the microbial As(V) and ferrihydrite-Fe(III) reduction kinetics respond to changes in the microbe-ferrihydrite suspension compositions in terms of cell densities and concentrations of As(V), ferrihydrite and lactate, (3) help understand the relationship of microbial ferrihydrite-Fe(III) and As(V) reduction and whether the priority between microbial ferrihydrite-Fe(III) and As(V) reduction can be explained by thermodynamics by ΔG_{rxn} (a Gibbs free energy change of a reaction).

2. Materials and methods

2.1. Synthesis of ferrihydrite

Ferrihydrite ($Fe_5HO_8 \cdot 4H_2O$) was prepared by slow addition (in droplets) of 1 M NaOH to 500 mL of a stirred solution of 0.2 M $FeCl_3$ until pH 7.0 was reached (Cornell and Schwertmann, 2003). The suspension was centrifuged (20 min, 2100 *g*, 20 °C) and washed free of excess salts with autoclaved doubly-deionised (DDI) water (>18 MΩ cm) from a Milli-Q system (Millipore, USA). The ferrihydrite was directly used for experiments without drying or freezing to prevent changes in particle aggregation and microporous structure. Ferric chloride instead of Fe(III) nitrate was used for ferrihydrite synthesis to avoid nitrate impurities that could serve as electron acceptor for microbial respiration. All beakers, bottles, serum bottles, and neutral salt solutions were autoclaved individually. Ferrihydrite was synthesised using the sterile solutions and glass containers and synthesis was carried out in a sterile bench. X-ray diffraction analysis confirmed the phase identity of ferrihydrite (2-line) (Huang et al., 2011b).

2.2. Microbial As(V) reduction experiments

Shewanella putrefaciens strain CN-32, obtained as ATCC® BAA-1097™, was grown for 24 h to late-exponential phase in tryptic soy broth at 30 °C in closed falcons with headspace initially filled with sterilised air (1:10 (v/v) air to solution ratio). The growth medium was slightly oxic (210 ± 16 mV) before cell culturing and became anoxic (−268 ± 27 mV) when cell harvesting. Cell densities of each culture were estimated from optical density measurements at 600 nm (OD_{600}), which were calibrated against total cell numbers determined by counting of DAPI (4′,6-diamidino-2-phenylindol)-stained cells using an optical microscope (Leica Microsystems, Wetzlar, Germany). Cells were harvested by centrifugation (2100 *g*, 15 min at 4 °C) and washed twice with a solution containing 10 mM PIPES buffer (pH 7.0) and neutral salts (5 mM NaCl; 0.5 mM $CaCl_2$; 0.5 mM $MgCl_2$). The washed cells were re-suspended in 2 mL wash solution and kept at 4 °C for less than 3 h before inoculation.

The kinetics of microbial reduction of As(V) was studied in batch experiments using 50 mL serum bottles capped with butyl rubber septa and crimp-seals. All experiments were carried out in 10 mM PIPES buffer adjusted to pH 7.0 and at constant background electrolyte concentrations (5.0 mM NaCl; 0.5 mM $CaCl_2$; 0.5 mM $MgCl_2$). As(V) and lactate were added from filter-sterilised solutions (0.2 μm). Initially, all solutions or suspensions contained 20–1000 μM As(V), 0.0935–93.5 mM ferrihydrite-Fe(III) or 2 mM citrate-Fe(III), 25 and 2500 μM Na-lactate, and ~$2.5 \times 10^9 - 1.5 \times 10^{10}$ cells mL^{-1}. First, the As(V) solutions or suspensions containing the sorbent minerals and all solutes were

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