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Role of Arsenic during the Aging of Acid Mine Drainage Precipitates

Pablo Cruz-Hernández^{a,1}, Rafael Pérez-López^a and José Miguel Nieto^a

^aDepartment of Geology, University of Huelva, Campus 'El Carmen' s/n, 21071, Huelva, Spain

Abstract

Iron-rich sediments cover the riverbeds affected by acid mine drainage (AMD) resulting from sulfide mineral oxidation. Precipitates are mainly composed of schwertmannite, which is a poorly-crystalline Fe-oxyhydroxysulfate that recrystallizes over a short-time period to goethite. Schwertmannite precipitation has a strong capacity for removal of some toxic elements like As. This study examines the influence of the initial As(V) concentration on the kinetics of precipitation and transformation of schwertmannite by means of batch experiments. A set of schwertmannites were synthesized with different As concentrations, and solid-solution interactions were allowed at 60°C during different time periods (from 1 h to 75 d). The increase of the initial As concentration notably the precipitation of schwertmannite and its transformation to goethite. Moreover, the transformation of schwertmannite into goethite entails the release of sulfate and, at a longer time scale, of part of the previously retained As. Thus, As acts as a retardant for schwertmannite transformation; however, this toxic element is released from the precipitates once the schwertmannite is transformed, which is *per se* an environmental paradox. Furthermore, schwertmannite precipitation also plays an important role in the AMD treatment systems; hence, the long-term behavior of these precipitates should be considered for the solid waste management.

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* Corresponding author. Tel.: +34-959-239-682; fax: +34-959-219-810.

E-mail address: Pablo.cruz@dgeo.uhu.es

1. Introduction

Arsenic is one of the elements considered as more harmful for the environment and human beings. This toxin is often associated with the metallic sulfide ore bodies. Sulfide oxidation under meteoric conditions leads to acidification of waters and release of high concentrations of sulfates and metal(oid)s such as Fe, As, Cd, Cu, Zn, and Pb. The oxidative leaching of sulfide-rich mining wastes is known as acid mine drainage (AMD). Under these conditions, a poorly-crystalline oxyhydroxysulfate, schwertmannite $[\text{Fe}_8\text{O}_8(\text{OH})_x(\text{SO}_4)_y \cdot n\text{H}_2\text{O}]$, $x=8-2y$, $1 < y < 1.75$, spontaneously precipitates on AMD-affected riverbeds^{1,2}. Schwertmannite is considered as an efficient scavenger of trace elements from the solution, showing predilection for As^{3,4}. However, this phase is metastable and transforms, under natural conditions, into goethite (FeOOH) in some weeks. During this short-term maturation, most of As remains linked to the solid phase⁵. Following the observations of Ford⁶ for ferrihydrite ($\text{Fe}_2\text{O}_3 \cdot 0.5\text{H}_2\text{O}$), it is expected that precipitation and transformation of schwertmannite can be also affected by As concentration in solution; although no information on this point has been found in the literature. Throughout this study, reaction rates of precipitation and transformation of schwertmannite were calculated in the laboratory using different As(V) concentrations in solution and heat to accelerate the transformation process.

2. Methodology

2.1. Sample preparation

The precursor phase, schwertmannite, was synthesized following the protocol of⁷ but in the presence of different As(V) concentrations and at 60°C. Thus, batch experiments were carried out by previously heating deionized water and adding 2 g L⁻¹ of pre-dehydrated $\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$ and different concentrations of $\text{Na}_2\text{HAsO}_4 \cdot x\text{H}_2\text{O}$ (0, 0.05, 0.0125, 0.25, 0.5 and 1 mM of As(V)). The solutions remained in the oven for variable time periods, ranging from 1 hour to 75 days. After experiments, solutions were filtered through 0.45 μm nylon syringe filters, acidified with suprapure HNO_3 and stored at 4°C until analysis. Solid samples were recovered by filtering solutions using cellulose nitrate membrane filters in a vacuum system coupled to a Büchner flask. These solids were rinsed twice with deionized water and dried at room temperature in presence of silica gel in order to avoid further mineral transformation.

2.2. Sample analysis

Concentrations of dissolved As, Fe, S, and Na in the solutions were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES; Jobin-Yvon Ultima2). The analytical errors were estimated to be below 5%. To check the accuracy of the results, the analysis sequence consisted of laboratory standards, quality control solutions analyzed as blind samples, blanks and duplicates. For solid characterization, bulk powder x-Ray diffraction (XRD) was performed by using a Bruker D8 advance diffractometer with $\text{CuK}\alpha$ radiation. Diffractometer was fitted at 40kV, 30 mA, and with a scan range of 2-65° 2θ, 0.05 °2θ step size, and 20 s time per step. All analyses were performed in the laboratories of the University of Huelva.

3. Results

3.1. Mineralogical identification

Powder XRD data show that schwertmannite was the only phase precipitated initially in most of the experiments. Only in experiments with the highest As concentration (1 mM) an amorphous phase was initially formed instead of schwertmannite. Secondly, schwertmannite transformed into goethite, which progressively appeared in all the batch experiments. The occurrence time of goethite increased with the initial As concentration; ranging from 48 h in the As-free experiment to 240 h in the experiment with 1 mM of As. This observation is consistent with those previously reported for ferrihydrite by⁶.

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