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Geomicrobiological investigation of two different mine waste tailings generating acid mine drainage

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Abstract

The impact of microbiological metal sulfide oxidation on acid mine drainage generation was studied for two different mine tailings. Microorganisms were quantified using different methods: (1) SYBR Green II direct counting, (2) TaqMan quantitative, real-time PCR (Q-PCR), (3) catalyzed reporter deposition–fluorescence in situ hybridization (CARD-FISH) and (4) most probable number (MPN) cultivation of acidophilic Fe(II) oxidizers. Potential pyrite or pyrrhotite oxidation rates were measured by microcalorimetry.

In the uncovered, pyrrhotite-containing tailings near Selebi-Phikwe, Botswana, acidophilic Fe(II)-oxidizing microorganisms were present in high numbers (MPN) of up to 10^7 cells g^{-1} dw (mean value 3×10^6 cells g^{-1} dw) throughout the entire water unsaturated, oxidized zone of about 25 m (at the tailings dam periphery) with a paste pH in the range of 3–4. Mean numbers of living Bacteria (CARD-FISH) and total microorganisms (SYBR Green II) were 1×10^7 cells g^{-1} dw and 8×10^7 cells g^{-1} dw, respectively. Cell numbers obtained by Q-PCR analysis were in the same range. The average potential pyrrhotite oxidation rate measured by microcalorimetry was 3.4×10^{-4} mol pyrrhotite m^{-3} tailings s^{-1} at 25 °C. About half of the pyrrhotite oxidation activity was biologically catalyzed.

By contrast, in the covered pyrite-containing tailings in Impoundment 1 in Kristineberg, northern Sweden, acidophilic Fe(II)-oxidizing microorganisms (mean value 5×10^5 cells g^{-1} dw) were only detected in a distinct zone of oxidized tailings between the cover and the unoxidized tailings where low pH values down to 3 prevailed. Bacterial numbers obtained by Q-PCR analysis were much higher (mean value 3×10^8 cells g^{-1} dw). The proportion of biological pyrite oxidation was up to 100% for the oxidized zone. The average potential pyrite oxidation rate was 1.6×10^{-5} mol pyrite m^{-3} tailings s^{-1} at 10 °C, an order of magnitude lower than that for the pyrrhotite-containing tailings.

Keywords: Acid mine drainage; Mine tailings; *Acidithiobacillus ferrooxidans*; Quantitative real-time PCR; CARD-FISH; SYBR Green II

1. Introduction

Acid mine drainage (AMD) is a strongly acidic solution containing high amounts of heavy metals and sulfate threatening groundwater quality. AMD is generated by chemical and biological oxidation of pyrite, pyrrhotite and other metal sulfides in mine waste heaps or in tailings from sulfidic ore processing [1–3]. The pyrite oxidation rate depends on temperature, pH, humidity and the availability of oxygen in the tailings, which is mainly controlled by diffusion. In addition, the oxidation rate strongly depends on the abundance of acidophilic Fe(II)- and metal sulfide-oxidizing microorganisms, which accelerate the kinetics of pyrite

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oxidation 30–300-fold [4,5]. Geomicrobiological and geochemical studies of pyrite and pyrrhotite-containing mine tailings pointed out the important role of acidophilic Fe(II)-oxidizing microorganisms, such as *Acidithiobacillus ferrooxidans* for the generation of AMD [6–11]. Only a few studies included the measurement of pyrite or pyrrhotite oxidation rates [9,12,13].

In mine waste, pyrite is oxidized according to Eq. (1):

$$FeS_2 + 3.5O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \quad (1)$$

Pyrrhotite oxidation occurs according to Eq. (2) [14]:

$$Fe_{1-x}S + (2-x/2)O_2 + xH_2O \rightarrow (1-x)Fe^{2+} + SO_4^{2-} + 2xH^+ \quad (2)$$

Based on the mineralogical composition of pyrrhotite, x ranging between 0 and 0.125 Fe(II) may be further oxidized to Fe(III) hydroxide according to Eq. (3):

$$Fe^{2+} + 0.25O_2 + 2.5H_2O \rightarrow Fe(OH)_3 + 2H^+ \quad (3)$$

In the present study, the uncovered pyrrhotite-containing tailings dam near Selebi-Phikwe, Botswana [15], and the covered pyrite-containing tailings in Impoundment 1 in Kristineberg, northern Sweden [16–21], were geomicrobiologically analyzed including the microcalorimetric measurement of potential pyrite oxidation rates [9,10,22,23]. The abundance of bacteria in the tailings has been quantified using four different techniques: (a) SYBR Green II staining, (b) TaqMan quantitative, real-time PCR (Q-PCR), (c) catalyzed reporter deposition–fluorescence in situ hybridization (CARD-FISH) and (d) most probable number (MPN) cultivation of acidophilic Fe(II)-oxidizers.

2. Materials and methods

2.1. Site description and sampling

2.1.1. Covered pyrite-containing tailings in Impoundment 1 in Kristineberg, northern Sweden

The annual precipitation in the humid Kristineberg area, northern Sweden, varies between 400 and 800 mm $year^{-1}$ and the annual mean temperature is 0.7 °C. The design, the mineralogy and the chemistry of the pyrite-containing tailings in Impoundment 1 have been described in detail [16–21]. Briefly, the tailings in Impoundment 1 cover an area of 0.1 km^2 and have a thickness of up to approximately 11 m, with an average thickness of 6–8 m. They were covered in 1996 with a soil cover consisting of 0.3-m compacted till and 1.5-m unspecified till. From the 1940s until 1996, the impoundment was unremediated and sulfide oxidation occurred in distinct depth layers (oxidized tailings). Based on the chemical composition, the sulfide mineral content of the unoxidized tailings ranges from 10% to 30%, totally dominated by pyrite. In the oxidized tailings, the sulfide content is generally lower [16]. Three boreholes at different locations (cores K, O and Q) were drilled using a drill-rig. The drill cores were split into 30 subsamples from the oxidized and the unoxidized tailings for laboratory analysis. Brown precipitates of iron (hydrox)oxides due to pyrite oxidation were found in the oxidized tailings at all three locations.

2.1.2. Uncovered pyrrhotite-containing tailings dam near Selebi-Phikwe, Botswana

The climate in Selebi-Phikwe, Botswana, is semiarid with an average annual temperature of 21 °C. The mine waste tailings dam near Selebi-Phikwe generates high amounts of AMD, which are collected in a drainage ditch surrounding the dam. To prevent contamination of surface water, AMD is purified in a plant by addition of limestone to increase the pH and to precipitate metals. The geochemistry of the tailings dam has been previously described [15]. Briefly, the tailings dam consists of waste from about 32 years of Ni-, Cu-, Zn- and Co-sulfidic ore processing. The original material from the flotation plant consists of about 35% solid material with an average grain size diameter of about 0.1 mm. The solid material contains about 11% pyrrhotite, about 1.5% other metal sulfides and hornblende and feldspar as major gangue minerals. The approximately 40-m high dam (2003) covers an area of ca. 1 km^2. The final height shall be 50 m in the year 2014. Currently, the surface of the central part is water covered, whereas the periphery of the dam surface is dry. Here, three holes (B-H1 = core 1, B-H2 = core 2, B-H3 = core 3) were drilled through the water unsaturated down to the saturated zone at about 25 m depth. Brown precipitates of iron (hydrox)oxides due to pyrrhotite oxidation were found throughout the entire unsaturated zone. A high proportion of the originally deposited pyrrhotite has been already oxidized within the first years of tailings deposition. Altogether, 65 solid samples were taken in 1-m intervals.

2.2. Geochemical analysis

The paste pH was measured with an electrode after shaking of 5-g sample in 12.5 mL 1 M KCl for 1 h. Humidity was determined as weight difference after