



Effect of neutralized solid waste generated in lime neutralization on the ferrous ion bio-oxidation process during acid mine drainage treatment



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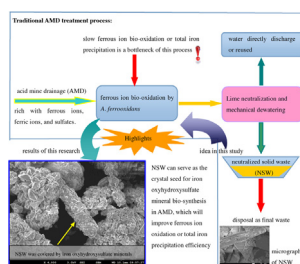
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HIGHLIGHTS

- Ferrous ion oxidation prior to lime neutralization is necessary for AMD treatment
- Neutralized solid waste (NSW) can enhance ferrous ion bio-oxidation efficiency
- NSW can act as seed for iron oxyhydroxysulfate mineral bio-synthesis
- NSW can improve total iron precipitation during ferrous ion bio-oxidation process

GRAPHICAL ABSTRACT



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ABSTRACT

Bio-oxidation of ferrous ions prior to lime neutralization exhibits great potential for acid mine drainage (AMD) treatment, while slow ferrous ion bio-oxidation or total iron precipitation is a bottleneck in this process. In this study, neutralized solid waste (NSW) harvested in an AMD lime neutralization procedure was added as a crystal seed in AMD for iron oxyhydroxysulfate bio-synthesis. The effect of this waste on ferrous ion oxidation efficiency, total iron precipitation efficiency, and iron oxyhydroxysulfate minerals yield during ferrous ion bio-oxidation by *Acidithiobacillus ferrooxidans* was investigated. Ferrous ion oxidation efficiency was greatly improved by adding NSW. After 72 h incubation, total iron precipitation efficiency in treatment with 24 g/L of NSW was 1.74–1.03 times higher than in treatment with 0–12 g/L of NSW. Compared with the conventional treatment system without added NSW, the iron oxyhydroxysulfate minerals yield was increased by approximately 21.2–80.9% when 3–24 g/L of NSW were added. Aside from NSW, jarosite and schwertmannite were the main precipitates during ferrous ion bio-oxidation with NSW addition. NSW can thus serve as the crystal seed for iron oxyhydroxysulfate mineral bio-synthesis in AMD, and improve ferrous ion oxidation and total iron precipitation efficiency significantly.

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1. Introduction

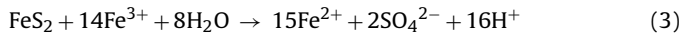
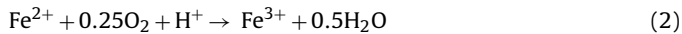
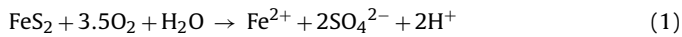
At present, the number of large and medium-sized coal mines in China exceeds 15,000. These produce more than 3.5×10^9 t coal per year, which accounts for more than 30% of the global coal production [1]. Some geological and environmental problems involved

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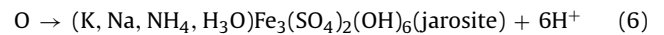
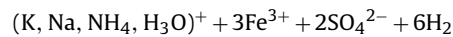
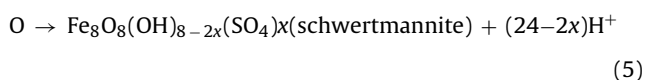
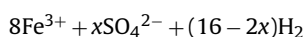
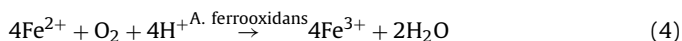
in geology, soil, water, and the atmosphere can arise due to coal mining activity, of which acidic mine drainage (AMD) is typical [2,3]. AMD mainly results from the oxidation of sulfide minerals, especially pyrite (FeS_2), by oxygen or the ferric ion when sulfide minerals are exposed to water and oxygen by coal mining activities [4,5]. Eqs. (1)–(3) describe the main chemical reactions:



AMD adversely affects the health of aquatic flora and fauna in surface water ecosystems through acidification and the contribution of considerable concentrations of iron and sulfates, and a trace amount of other soluble toxic metals (e.g., Zn, Al, Mn, Pb) or metalloids (e.g., As and Hg) [6–9]. For example, the AMD in Mina Esperanza, located in the northern part of the Iberian Pyrite Belt, in Southwestern Spain, contains 755–1100 and 3324–4515 mg/L of iron ions and sulfate ions, respectively; however, it contains just 0.36–0.69, 0.07–0.10, 0.47–0.76, 0.02–0.04, and 0.15–0.25 mg/L of arsenic, cadmium, cobalt, chromium, and nickel, respectively [10]. AMD could reduce the biological diversity in streams [11]. Furthermore, the yellow-orange iron hydroxide precipitate caused by high concentrations of iron in AMD will increase the concentration of suspended solids and prevent light penetration into the subsequent benthic layer in AMD receiving waterways, thereby impairing the photosynthetic capacity of autotrophic organisms [7,8]. Considering these environmental and ecological threats, an effective technology is needed to decrease the acidity of AMD, as well as the concentration of iron ions, sulfates, and trace heavy metals therein, and to facilitate water reuse.

Several technologies have been used to treat AMD including chemical neutralization by alkaline substances, microbial treatment by sulfate reducing bacteria, wetlands technology, and ion exchange membrane methods [12–15]. A commonly used chemical method for treating AMD is lime neutralization, which raises the pH and removes the iron and sulfate, mainly through the formation of ferric hydroxide and calcium sulfate [12,16]. Oxidation of ferrous ions to ferric ions is required prior to lime neutralization since ferric ions could precipitate at a much lower pH than ferrous ions [17]. Oxidation of ferrous ions has been achieved by aeration traditionally [12]; however, the abiotic oxidation of ferrous ions by aeration is very slow at pH below 4.0. Therefore, developing technologies for improving the oxidation efficiency of ferrous ions under low pH is of paramount significance in AMD treatment.

A promising approach for AMD treatment using acidophilic microorganisms was designed at the Nochten opencast pit in Lusatia, Germany [3]. In this process, ferrous ion oxidation is catalyzed by acidophilic microorganisms, such as *Acidithiobacillus ferrooxidans* (*A. ferrooxidans*), and ferrous ions can be oxidized easily at pH of approximately 2–4 [18]. It is noted that *A. ferrooxidans* can accelerate the ferrous ion oxidation process by 10^5 to 10^6 times [19]. Therefore, rapid bio-oxidation of ferrous ions by *A. ferrooxidans* has great potential as a pre-treatment process prior to lime neutralization for AMD treatment. Iron oxyhydroxysulfate minerals, such as schwertmannite or jarosite, are generated during ferrous ion oxidation by *A. ferrooxidans*; ferric ions is facilitated [20,21]. Eqs. (4)–(6) describe schwertmannite or jarosite precipitation [21,22]:



The formation of iron oxyhydroxysulfate minerals during ferrous ion bio-oxidation is effective in removing iron from AMD, thereby reducing the amount of lime required and improving the reaction efficiency in any subsequent AMD lime neutralization process [17]. Furthermore, iron oxyhydroxysulfate minerals have a great capacity for scavenging toxic heavy metals, such as Pb and As, through adsorption and co-precipitation [23,24], which will remove a portion of these elements from solution. In addition, compared with ferric hydroxide, iron oxyhydroxysulfate minerals possess better settling characteristics [25], which aid the dewatering of neutralized solid waste (NSW) after AMD treatment. Therefore, iron oxyhydroxysulfate minerals efficiently synthesized during ferrous ion oxidation by *A. ferrooxidans* have great significance for AMD treatment.

“Seeding” is the most desirable means by which to promote jarosite formation [26,27]. Dutrizac [26] found that during the jarosite chemical synthesis process, the jarosite precipitation rate seems to increase in a linear manner with the amount of jarosite seed present. Song et al. [27] demonstrated that the extra jarosite seed addition can enhance jarosite precipitation efficiency during bio-oxidation of ferrous ions by *A. ferrooxidans*. Other crystalline minerals, such as silica sand, also could play the role of seed during the jarosite bio-synthesis process [28]. For example, Wang and Zhou [28] found that adding 40 g/L of silica sand as the seed in jarosite bio-synthesis facilitated by *A. ferrooxidans* increased the total iron precipitation rate by 24%. A mixture of calcium sulfate and ferric hydroxide is generated as the main NSW during AMD lime neutralization after ferrous ion bio-oxidation, and calcium sulfate is a typical crystalline mineral. However, it is not yet clear whether NSW can act as the seed during iron oxyhydroxysulfate minerals formation to improve total iron precipitation efficiency. If NSW can serve as a seed, it would be a useful material to improve the AMD treatment efficiency, rather than simply a waste requiring further management. In addition, little information is available on the bio-oxidation efficiencies of ferrous ions during iron oxyhydroxysulfate mineral bio-synthesis by *A. ferrooxidans* with seed addition.

The objectives of the present study were: (1) to investigate the effect of NSW as seed on bio-synthesis of iron oxyhydroxysulfate minerals during ferrous ion bio-oxidation by *A. ferrooxidans* and (2) to examine the oxidation efficiency of ferrous ions during its bio-oxidation in the presence of NSW added as seed in AMD treatment.

2. Materials and methods

2.1. *A. ferrooxidans* and its culture

The strain of *A. ferrooxidans* LX5 (CGMCC No.0727) used in this study was obtained from the China General Microbiological Culture Collection Center (CGMCC) and was grown in modified 9K liquor medium [29]. The modified 9K medium contained the following analytical grade salts: 0.0168 g of $\text{Ca}(\text{NO}_3)_2$, 0.058 g of K_2HPO_4 , 0.119 g of KCl, 0.583 g of $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 3.5 g of $(\text{NH}_4)_2\text{SO}_4$, and 44.2 g of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ in 1 L of deionized H_2O , adjusted to pH 2.50 with 9 mol L^{-1} H_2SO_4 . The $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution was sterilized by filtration through 0.22 μm filters, and then added to the other autoclaved (at 121 °C for 15 min) medium components [30]. In this study, inorganic salt concentration of modified 9K medium was increased ten times for the prepared modified 9K liquor medium stock solution before experiment.

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