



Application of ferrate for the treatment of metal-sulfide

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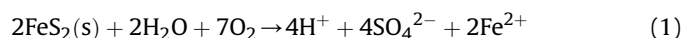
ABSTRACT

Fe(VI) was evaluated to treat metal-sulfides such as Fe–S, Pb–S, Cu–S and Cd–S contained in mine tailings known to generate acidic mine drainages. The rate of Fe(VI) reduction was dependent on the type of metal-sulfide as well as the concentration of each metal-sulfide. Fe(VI) reduction increased as the concentration of each metal-sulfide increased. The order of initial rates for the Fe(VI) reduction was Pb–S > Cu–S > Fe–S > Cd–S. The rate of Fe(VI) reduction by each metal sulfide increased as the ionic strength increased. For all metal sulfides, reduction efficiency of Fe(VI) was not affected by the presence of different background electrolytes except NaNO₂ and Na₂SO₃. This result suggests that fully oxidized anions such as ClO₄[−], NO₃[−], SO₄^{2−} as well as redox insensitive anion such as Cl[−] are not involved in the redox reaction between Fe(VI) and metal sulfides.

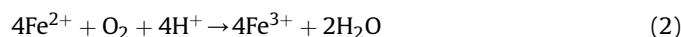
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1. Introduction

Acid mine drainage (AMD) is generated from the oxidation of exposed mine tailings containing metal sulfides. When sulfides are exposed to air and water, sulfuric acid is produced through several chemical reactions. In particular, pyrite produces ferrous ions and sulfuric acid through chemical and biological reactions (Bigham et al., 1990; Chon et al., 1999; Hashim et al., 2011; Sharma, 2011; Stumm and Morgan, 1996). Chemical oxidation of pyrite can be expressed as follows:



In Eq. (1), sulfide is oxidized to form H⁺ and SO₄^{2−}. The liberated soluble Fe²⁺ is also oxidized to ferric ion:



Therefore acid mine drainage normally contains high levels of metal and inorganic ions such as iron, aluminum, and sulfate. AMD often has an orange or yellowish-orange color, indicating high levels of iron, and may also have the smell of rotten egg due to the release of hydrogen sulfide. Contamination of river and ground water by AMD cause several environmental problems such as

putting at risk drinking water sources and ecosystem damages (Bilek and Wagner, 2012; Costa et al., 2009; Silveira et al., 2009).

Since mine tailings and waste rocks, which contain metal sulfides, are a significant source of AMD, proper management of these waste dumps is highly necessary. Neutralization and precipitation have been widely applied as general processes for the treatment of mine drainage containing high levels of metal ions (Akcil and Soner, 2006). However it is known that accurate prediction of the pH of spoil run-off is difficult due to the self-neutralizing character of carbonate-containing spoils (Akcil and Soner, 2006). In addition, these processes are end-of-pipe. Therefore pre-treatment is regarded as an important process to reduce or eliminate the effects of AMD.

As a promising pre-treatment process, ferrate, Fe(VI), the higher oxidation state of iron, can be applied to treat mine tailing or waste rocks because it possesses a very high oxidative capacity (redox potential, E°, is 2.20 and 0.72 V at pH 1 and 14, respectively), and the Fe(III) generated by the reduction of Fe(VI) is much less harmful toward human beings or the environment; Fe(III) also readily forms precipitates (Tiwari et al., 2005). An additional advantage of using Fe(VI) in remediation processes is that it can be considered a green chemical (Sharma, 2002, 2004; Sharma et al., 2005).

However there is very limited information on the applicability of Fe(VI) for the treatment of mine tailings and waste rocks containing various metal sulfides. Murshed et al. (2003) used Fe(VI) to treat sulfide mine tailing through oxidation process. However, comparative studies on the oxidation of sulfide from each metal

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sulfide is highly necessary to provide fundamental data to optimize the application of Fe(VI) in the treatment of real mine tailings or waste rock.

Thus, in this study, oxidation efficiency, oxidation kinetics, and oxidation pattern of sulfide for several metal-sulfides using Fe(VI) was investigated at various solution pH, concentrations of metal-sulfide, ionic strength, and types of background electrolytes.

2. Materials and methods

2.1. Chemicals used

Fe–S, Pb–S, Cu–S and Cd–S: GR grade were obtained from Aldrich, USA. Sodium hypochlorite, purified Fe(III) nitrate hexahydrate, potassium hydroxide, *n*-hexane, anhydrous ether and methanol were obtained from Merck. The deionized water (18 M Ω cm) used for sample preparation and all experiments was obtained using a Milli Q-Plus Instrument (Millipore SA 67120, Molsheim, France). A stock solution (1×10^{-2} mol/L) of Fe(VI) was prepared with deionized water and used for further experiments with dilution. All other chemicals used were AR/GR grade unless otherwise mentioned.

2.2. Preparation of potassium ferrate

Potassium ferrate was prepared by a wet chemical oxidation method, with some modifications as described in previous studies

(Li et al., 2004; Tiwari et al., 2007). In this method, sodium hypochlorite (12–14%) was used as an oxidant. In a beaker, 90 g of solid KOH were added slowly to 300 mL of chilled NaClO solution. After cooling the resulting suspension, the precipitate formed was filtered with GF/C filter paper. The filtrate exhibited a clear yellow color and was a highly alkaline NaClO solution. Again this alkaline NaClO solution was chilled and filtered using a GF/C filter paper to remove any additional precipitates formed within the solution. Within 30 min, 20 g of pulverized ferric nitrate were added slowly into this solution, with constant and vigorous stirring under cooling conditions (<8 °C) because oxidation of Fe(III) to Fe(VI) is favorable at lower temperatures. After complete addition of the ferric nitrate, the solution was stirred for another 30 min to enhance yield. In this step, the color of the solution readily changed to purple. Next, approximately 50 g of solid KOH was added while maintaining the solution temperature below 15 °C to avoid decomposition of Fe(VI). The solution mixture was allowed to cool in a refrigerator for 60 min. The resulting dark purple slurry was filtered with a glass filter (medium porosity 10–15 μ m), and the filtrate was discarded. Then the precipitate was washed with 100 mL of chilled 3 M KOH solution. The filtrate from this washing process was collected, placed in a beaker and 100 mL of saturated chilled KOH solution was then added. The potassium ferrate readily precipitated, and was filtered again with a GF/C filter. The filtrate was discarded, and the solid was washed with 50 mL of cold 3 M KOH solution. Re-precipitation was carried out at least for 3–4 times to remove any impurities. Finally, the solid was flushed with *n*-hexane (four

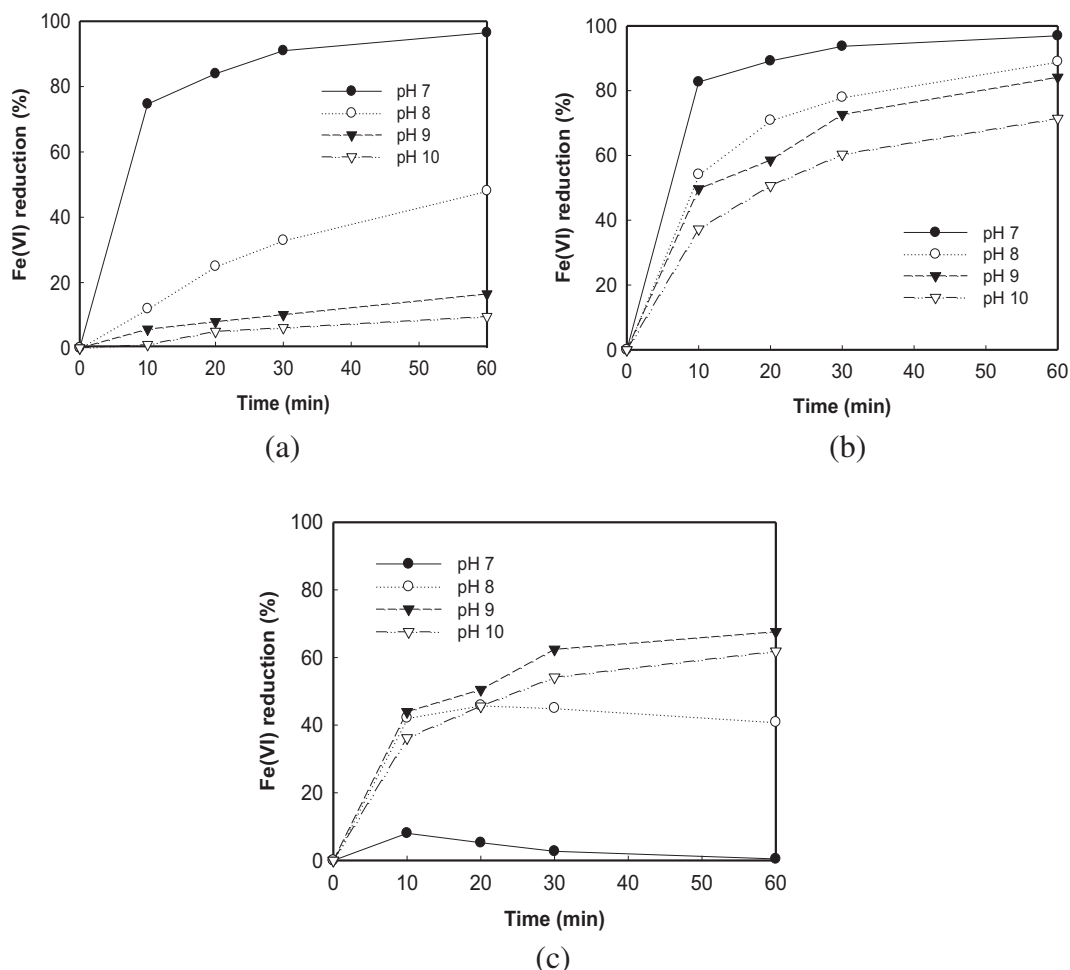


Fig. 1. Percent Fe(VI) removal (a) in the absence of Fe–S, (b) in the presence of Fe–S and (c) net Fe(VI) reduction as a function of time.

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