



Geochemical investigation of the galvanic effects during oxidation of pyrite and base-metals sulfides



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HIGHLIGHTS

- A geochemical investigation of galvanic interactions is proposed.
- Pyrite is galvanically protected upon contact with chalcopyrite or sphalerite.
- Sphalerite and chalcopyrite show higher reactivity rates in the presence of pyrite.

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ABSTRACT

Predicting the water quality at mine sites is of significant importance for developing mines with respect for the environment. Acid mine drainage (AMD) occurs when sulfides are in contact with oxygen and water, and several parameters and mechanisms influence final drainage quality. Galvanic interactions influence the reactivity of sulfide minerals, which act as semi-conductors. These galvanic interactions have been insufficiently studied in the context of AMD generation. In this study, the influence of pyrite on the reactivity of sphalerite and chalcopyrite was investigated. Five blends, comprised of free grains of quartz/pyrite, quartz/chalcopyrite, quartz/sphalerite, quartz/pyrite/chalcopyrite, and quartz/pyrite/sphalerite, were subjected to geochemical testing. Five weathering cells were monitored over a 200-day period during which they were leached twice weekly. Leachates were analyzed for pH, Eh, electrical conductivity, and sulfate and metal concentrations. The results of these analyses showed that galvanic interactions occurred between free sulfide grains. Pyrite was galvanically protected over the full testing period in the quartz/pyrite/chalcopyrite blend, and partially protected in the quartz/pyrite/sphalerite blend. Moreover, the release of Cu from chalcopyrite and Zn, Mn, and Cd from sphalerite was accelerated in the presence of pyrite. This work provides a better understanding of the influence of pyrite on chalcopyrite and sphalerite reactivity by highlighting the galvanic effects. In the future, to improve the reliability of AMD prediction tests, galvanic interactions should be considered in both the prediction of the acid generation potential and the estimation of metal and metalloid release rates.

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1. Introduction

The mining industry contributes greatly to the economic strength of many countries around the world, including Canada. In 2013, the extraction of minerals (metals, non-metals, coal)

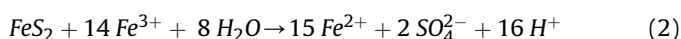
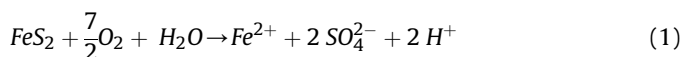
contributed \$22 billion to Canada's GDP (Marshall, 2014). Although Canada is a country of vast natural resources and natural habitats, including large forests and lakes, it is impacted by past practices and pollution caused by mineral extraction. Indeed, mining generates large amounts of liquid and solid wastes which must be managed properly to safeguard, maintain, and restore the environment. Currently, these wastes are mostly stored at the surface of mine sites. They may also generate contaminated mine drainage due to their exposure to air and water. Acid mine drainage (AMD) is

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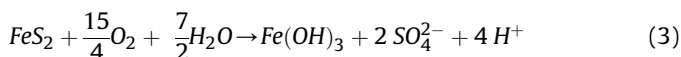
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the most well-known and problematic type of drainage facing the mining industry. Other types of drainage, such as contaminated neutral drainage (CND), saline drainage, or alkaline drainage can also occur. As these drainages can have a significant impact on the environment, these phenomena must be predicted during the feasibility stage by mining companies primarily to meet environmental regulations such as those required by an environmental impact assessment (EIA) study.

AMD occurs when a reactive mineral is exposed to an oxidant and water, either in oxygenated or anoxic systems, depending on the oxidant. Reactive minerals are sulfides and sulfosalts which are largely present in coal and metal mines. The processes leading to AMD are numerous and complex and involve chemical, biological, and electrochemical reactions. The chemical oxidation of sulfides can follow a variety of pathways involving surface interactions with dissolved O_2 , Fe^{3+} , and other mineral catalysts (e.g., MnO_2) (Blowes et al., 2014). Equation (1) presents the oxidation reaction of pyrite by O_2 at near-neutral pH, and Equation (2) by ferric iron Fe^{3+} at acidic pH under abiotic conditions.



The rate of pyrite oxidation by Fe^{3+} is faster than by O_2 (Mylona et al., 2000). In addition, the Fe^{2+} generated in Equation (1) can oxidize into Fe^{3+} and hydrolyze, which leads to the overall pyrite oxidation reaction presented in Equation (3).



The catalysis of AMD reactions by bacterial activity is well-known. Thus, biological reactions and associated microorganisms have been increasingly studied in mine environments. Recently, Nordstrom et al. (2015) highlighted advances in the knowledge of microbial activity. For example, microbial activity can increase the oxidation rate of chalcopyrite by up to three orders of magnitude with respect to purely chemical weathering (Kwong et al., 2003), and can significantly enhance zinc dissolution in sphalerite (Natarajan 1992).

Electrochemical reactions also play a significant role in the rates of oxidation of sulfides. Holmes and Crundwell (2000) showed that the electrochemical reaction steps occurring at the mineral – solution interface control the rate of dissolution of sulfides. More recently, Crundwell (2013) discussed the dissolution and leaching of minerals and concluded that the transfer of electrons at the mineral surface during leaching reactions controls the dissolution rate of minerals. These electrochemical reactions are responsible for the galvanic effects, which have been largely investigated in hydrometallurgy.

Galvanic reactions occur between conductive or semi-conductive minerals in aqueous systems. They play an important role in the aqueous processing of ores and minerals, such as in flotation and leaching (Buckley et al., 1989). For semi-conductive minerals, such as sulfides, direct contact of minerals with different rest potentials initiates the galvanic effect. This effect has been modeled with galvanic cells through redox reactions, where the mineral with the highest rest potential acts as the cathode, which is galvanically protected, while the mineral with the lowest rest potential acts as an anode and its dissolution is favored through electronic interactions. Galvanic interactions are a function of the mineralogical associations between the phases present in a material. Cruz et al. (2005) showed that even small inclusions of other minerals in pyrite can cause a significant modification of its

reactivity. Moreover, the rest potential of a mineral can vary, to a limited extent, depending on the sulfide's detailed composition (Shuey, 2012). In addition to the absolute difference in electrode potential between two contacting sulfides, the relative surface areas of the galvanic couple greatly influence the rate of oxidative dissolution of the anode sulfide because of the resultant current density generated. The larger the anodic area, the more widely distributed is the current generated by the galvanic cell; thus, the current density becomes lower. Low current density results in a slow dissolution of the anode sulfide. If the anode sulfide occurs as a minor inclusion in the cathode sulfide, its dissolution rate is increased due to the greater current density generated (Kwong et al., 2003).

In order to determine the galvanic interactions in a complex mineral association, strategies like cyclic voltammetry were suggested (Cruz et al., 2005). Mehta and Murr (1983) explained the phenomenon of galvanic interactions in terms of the rest potential of each mineral with respect to the standard hydrogen electrode (SHE). The authors examined a number of galvanic couples, including chalcopyrite and sphalerite in electrical contact with pyrite. It was observed that as the amount of pyrite in contact with another mineral increases, the rate of leaching of this mineral increases. Although galvanic interaction is a well-known phenomenon, the exact processes and mechanisms occurring are not thoroughly understood in sulfide mineral systems. Research to describe galvanic interactions in sulfide mineral systems is primarily qualitative. Hepel (1984) investigated the mechanisms which lead to selective leaching in sulfide mineral systems and observed that galvanic cell formation between electrically conducting minerals hindered the leaching of some sulfides (Holmes and Crundwell, 1995). Galvanic interactions could also lead to preferential leaching of metals. Recent advances in hydrometallurgy have taken full advantage of this preferential leaching of metals by galvanic interactions among contacting sulfides to enhance metal extraction, especially base metals, both *in situ* and in mineral processing plants (Natarajan 1992).

More recently, galvanic sulfide oxidation has also been investigated from an environmental perspective (Kwong, 1995; Kwong et al., 1997; Kalinnikov et al., 2001; Kwong, 2001). However, the concept of galvanic sulfide oxidation as an important and natural metal leaching mechanism has not been widely accepted (Kwong et al., 2003). Cruz et al. (2005) showed that the occurrence of AMD can be delayed due to cathodic protection of an acid-generating sulfide such as pyrite. Moreover, water pollution from galvanic interactions can continue to occur for a long period of time, even after mining closure (Benvenuti et al., 1997). Some authors believe that the oxidative dissolution of sulfides through galvanic interactions is one of the most important factors that lead to environmental pollution in metal sulfide mine areas (Byerley and Scharer, 1992; Subrahmanyam and Forssberg, 1993; Salomons, 1995; Lin, 1997; Cruz et al., 2001; Liu et al., 2008).

Despite the complexity of sulfide oxidation and the possible influence of galvanic interactions, AMD prediction tests are still widely based on oversimplifications of AMD generating processes. This may lead to mismanagement of tailings and waste rocks and result in significant environmental problems and added costs. For AMD prediction, static tests are used as a screening tool to predict if a material will generate acid or not. These predictive tests are based only on basic calculations from chemical assay data or on the proportions of different mineralogical phases present in a material (Bouzahzah et al., 2014; Hageman et al., 2015).

The main objective of this study was to investigate the influence of coupling sulfide minerals on their respective reaction rates. Five kinetic tests (weathering cells) were set up with three common sulfides. Two sulfide mixtures were prepared and consisted of

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