



## Suppressive effects of ferric-catecholate complexes on pyrite oxidation

Xinlong Li <sup>a,\*</sup>, Naoki Hiroyoshi <sup>b</sup>, Carlito Baltazar Tabelin <sup>b</sup>, Kosuke Naruwa <sup>c</sup>,  
Chie Harada <sup>c</sup>, Mayumi Ito <sup>b</sup>

<sup>a</sup> Laboratory of Mineral Processing and Resources Recycling, Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan

<sup>b</sup> Laboratory of Mineral Processing and Resources Recycling, Division of Sustainable Resources Engineering, Faculty of Engineering, Hokkaido University, Japan

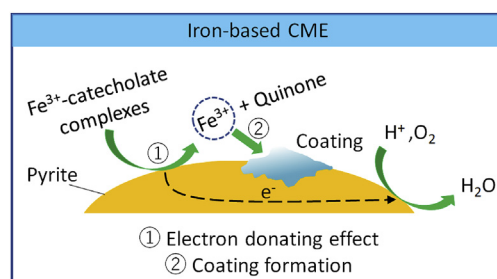
<sup>c</sup> Laboratory of Mineral Processing and Resources Recycling, Division of Cooperative Program for Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan



### HIGHLIGHTS

- Fe<sup>3+</sup>-catecholate complexes were oxidatively decomposed to release Fe<sup>3+</sup> on pyrite surface.
- Fe<sup>3+</sup> was precipitated as an iron oxyhydroxide coating on pyrite surface.
- Iron-based CME suppressed pyrite oxidation and could depress its floatability in mineral processing.

### GRAPHICAL ABSTRACT



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### ABSTRACT :

Pyrite, a common gangue mineral in complex sulfide ores and coals, is rapidly oxidized in water by ferric ions and dissolved oxygen to form a very acidic and heavy metal-laden leachate called acid mine drainage (AMD). Carrier-microencapsulation (CME) using Ti<sup>4+</sup>, Si<sup>4+</sup>, and Al<sup>3+</sup> was reported as a promising new approach to prevent pyrite oxidation by forming a passivating barrier on the pyrite surface. In CME, the presence of Fe<sup>3+</sup>-catecholate complexes is unavoidable but their effects on pyrite oxidation remain unclear.

In this study, the effects of Fe<sup>3+</sup>-catecholate complexes on pyrite oxidation were investigated. Formations of mono-, bis-, and tris-catecholate complexes of Fe<sup>3+</sup> were verified by UV–Vis spectrophotometry and their speciation with pH was consistent with thermodynamic considerations. Linear sweep voltammetry was conducted to evaluate the redox properties of Fe<sup>3+</sup>-catecholate complexes, and the results indicate that ligands in the three complexes were sequentially oxidized until Fe<sup>3+</sup> is released. Coating formation on pyrite was confirmed after treatment with mono- and bis-catecholate complexes. Results of SEM-EDX and ATR-FTIR indicate that the coating is composed primarily of iron oxyhydroxide phases. The results of leaching experiments showed that pyrite oxidation was suppressed by Fe<sup>3+</sup>-catecholate complexes via two mechanisms: (1) electron donating effects of the complexes, and (2) formation of a protective coating on pyrite. The results provide not only a better understanding of the effects of Fe<sup>3+</sup>-catecholate complexes on pyrite oxidation but also some possible applications of Fe<sup>3+</sup>-based CME

\* Corresponding author. Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Kita 13, Nishi 8, Kita-ku, Sapporo, 060-8628, Hokkaido, Japan.

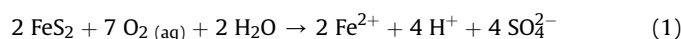
E-mail address: [rikinryu@gmail.com](mailto:rikinryu@gmail.com) (X. Li).

such as the suppression of pyrite oxidation to prevent AMD formation and depression of pyrite floatability in mineral processing.

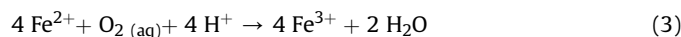
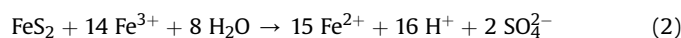
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## 1. Introduction

Pyrite (FeS<sub>2</sub>) is the most abundant metal sulfide mineral in nature (Bonnissel-Gissing et al., 1998), and is often associated with mineralized zones containing valuable metals such as gold, silver, copper, lead, and zinc as well as with coal deposits (Chandra and Gerson, 2010; Monterroso and Macias, 1998; Tabelin and Igarashi, 2009; Tabelin et al., 2012a). Pyrite is commonly separated from valuable minerals by mineral processing techniques and subsequently disposed of in tailings dams as wastes. Upon exposure to oxygen and water, pyrite is rapidly oxidized to form sulfuric acid according to the following reaction (Biegler and Swift, 1979; Holmes and Crundwell, 2000; Johnson and Hallberg, 2005; Todd et al., 2003):



Pyrite dissolution is further promoted in the presence of ferric ions (Fe<sup>3+</sup>), a strong oxidant of pyrite, which is produced from the oxidation of ferrous ions (Fe<sup>2+</sup>) by O<sub>2</sub> and/or the mediation of iron-oxidizing bacteria (Eqs. (2) and (3)) (Garrels and Thompson, 1960; Singer and Stumm, 1970; Plaza et al., 2017).



Sulfuric acid generated by pyrite oxidation could further enhance the dissolution of other minerals containing hazardous elements such as cadmium (Cd), arsenic (As), and lead (Pb) in the wastes (Jha et al., 2008; Tabelin et al., 2012b, 2017a; 2017b, 2017c; 2017d) and finally form a heavy metal-laden polluted acidic leachate called acid mine drainage (AMD). AMD is a serious environmental problem not only in the mining industry (Yuniati et al., 2015) but also in underground space development projects (e.g. roads and railway tunnels) (Tabelin et al., 2010, 2014a; 2014b; Tamoto et al., 2015; Tatsuhara et al., 2012). It is also a potential threat to surrounding ecosystems and the health of people living around AMD-polluted areas (Motsi et al., 2009; Simón et al., 2001; Tabelin et al., 2018; Yuniati et al., 2015). Once AMD is produced, its treatment by techniques like chemical neutralization is essential and has to be continued for long periods of time (even after the closure of mines) (Mackie and Walsh, 2012). If pyrite oxidation could be suppressed, AMD treatment costs would be reduced and a more sustainable approach to manage this environmental problem could be developed.

Several authors have proposed a new and promising approach to suppress the oxidation of sulfide minerals like pyrite and arsenopyrite by directly passivating their surfaces in a process called carrier-microencapsulation (CME) (Jha et al., 2008, 2011; Park et al., 2018a, 2018b; Satur et al., 2007; Yuniati et al., 2015). In this technique, metal (or metalloid) ions with low solubility are stabilized in the aqueous phase by forming complexes with a carrier (i.e., organic ligand). On the surface of sulfide minerals, the complex is oxidatively decomposed releasing the metal (or metalloid) ions that are precipitated as oxide or hydroxide. These precipitates

eventually cover the mineral surface forming a protective coating that inhibits contact between the oxidants and sulfide minerals, resulting in the suppression of their oxidations. The primary advantage of CME over other encapsulation techniques is its ability to specifically target pyrite and arsenopyrite in complex systems like mine waste and tailings, and several authors reported that CME using Ti<sup>4+</sup>-, Si<sup>4+</sup>-, and Al<sup>3+</sup>-catecholate complexes are effective in suppressing pyrite and arsenopyrite oxidations (Belzile et al., 1997; Jha et al., 2008, 2011; Park et al., 2018a; b; Satur et al., 2007).

When the application of CME in actual mine wastes and tailings is considered, it is important to know how coexisting minerals and metal ions affect pyrite oxidation and the extent of coating formation. The most common and important coexisting metal ions/minerals are the ions of iron (Fe<sup>3+</sup> and Fe<sup>2+</sup>) and its oxyhydroxides/oxides. For example, iron-oxide-copper-gold ore deposits (IOCG), which are important copper sources, contain magnetite (Fe<sub>3</sub>O<sub>4</sub>) as the dominant Fe-bearing mineral together with pyrite and chalcopyrite (CuFeS<sub>2</sub>). In tailings dams, a significant amount of Fe<sup>3+</sup> and Fe<sup>2+</sup> exist in the aqueous phase together with iron-bearing secondary minerals like hematite, goethite, akaganeite, and schwertmannite (Durocher and Schindler, 2011). Several authors have reported that Fe<sup>3+</sup> and Fe<sup>2+</sup> could form water-soluble complexes with catechol (Avdeef et al., 1978; Hider et al., 1981, 1983; Schweigert et al., 2001; Sever and Wilker, 2004) and catechol has the ability to extract Fe from its oxyhydroxide/oxide minerals (Gulley-Stahl et al., 2010). Therefore, Fe<sup>3+</sup>-catecholate complexes would be formed when CME is applied in the actual mining operations. The interactions of catechol with the ions of Fe and its oxyhydroxides/oxides are also important in the mobility of hazardous and toxic contaminants in the environment. This is primarily because catechol is a ubiquitous organic molecule present in natural organic matter while Fe-oxyhydroxides/oxides are the most important adsorbents of toxic metalloids like arsenic and selenium (Korshin et al., 1997; Tabelin et al., 2012a, 2012b; 2013, 2014c).

This study aims to investigate the effects of Fe<sup>3+</sup>-catecholate complexes on pyrite oxidation. A speciation diagram of Fe<sup>3+</sup>-catecholate complexes was created using hyperquad simulation and speciation software (Hyss, 2009) based on thermodynamic considerations and the reliability of the results was verified by ultraviolet–visible (UV–Vis) light spectrophotometry. Linear sweep voltammetry (LSV) was conducted to elucidate the redox properties of Fe<sup>3+</sup>-catecholate complexes. The composition and morphology of the pyrite surface treated with Fe<sup>3+</sup>-catecholate complexes were characterized by high-resolution optical microscopy, scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX) and attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy. Finally, the effects of Fe<sup>3+</sup>-catecholate complexes on pyrite oxidation were investigated by batch-type leaching experiments.

## 2. Materials and methods

### 2.1. Pyrite sample preparation and characterization

A pyrite specimen obtained from Cerro de Pasco Mine, Peru was used in this study. For surface analysis of oxidation products, a

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