



Simultaneous suppression of acid mine drainage formation and arsenic release by Carrier-microencapsulation using aluminum-catecholate complexes

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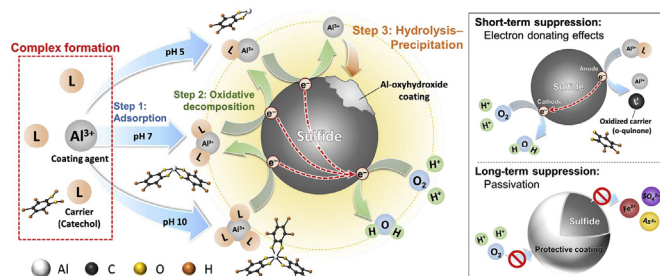
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HIGHLIGHTS

- Al-catecholate complexes were oxidatively and sequentially decomposed.
- [Al(cat)]⁺ was the most effective in suppressing pyrite and arsenopyrite oxidations.
- Minerals were covered with an Al-oxyhydroxide coating after Al-based CME.
- Coating was stable and inhibited the anodic half-cell reaction.
- Mechanism of Al-based CME involves adsorption, decomposition and coating formation.

GRAPHICAL ABSTRACT



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ABSTRACT

Pyrite (FeS₂), the most common sulfide mineral in nature, plays an important role in the formation of acid mine drainage (AMD), one of the most serious environmental problems after the closure of mines and mineral processing operations. Likewise, arsenopyrite (FeAsS) is an important sulfide mineral because its dissolution releases toxic arsenic (As) into the environment. To mitigate the serious environmental problems caused by pyrite and arsenopyrite, this study investigated carrier-microencapsulation (CME) using Al-catecholate complexes, a technique that selectively forms protective coatings on the surfaces of sulfide minerals, by electrochemical techniques and batch leaching experiments coupled with surface sensitive characterization techniques.

Cyclic voltammetry (CV) of Al-catecholate complexes (mono-, bis-, tris-catecholate) suggest that these three species could be oxidatively decomposed in this order: [Al(cat)₃]³⁻ → [Al(cat)₂]⁻ → [Al(cat)]⁺ → Al³⁺, and these reactions were irreversible. Among these three species, [Al(cat)]⁺ was the most effective in suppressing pyrite and arsenopyrite oxidations because it requires less steps for complete decomposition than the other two complexes. Analyses of CME treated minerals by scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX) and diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) indicated that they were covered with Al-oxyhydroxide (γ-AlO(OH)), which

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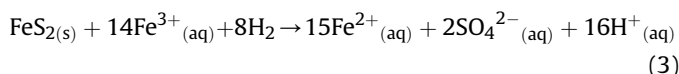
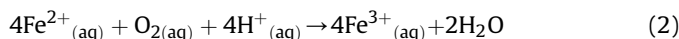
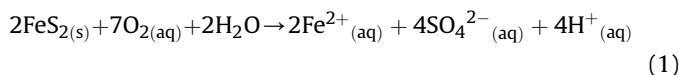
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became more extensive at higher $[\text{Al}(\text{cat})]^+$ concentrations. In addition, this coating was stable even at relatively high applied potentials that simulated surface oxidizing conditions. Based on these results, a detailed mechanism of Al-based CME is proposed: (1) adsorption of $[\text{Al}(\text{cat})]^+$ on the surface of mineral, (2) oxidative decomposition of $[\text{Al}(\text{cat})]^+$ and release of “free” Al^{3+} , and (3) precipitation and formation of Al-oxyhydroxide coating.

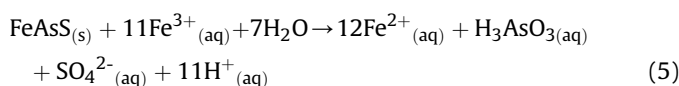
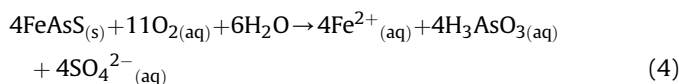
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1. Introduction

Pyrite (FeS_2), the most common sulfide mineral in nature, plays an important role in the redox cycling of iron (Fe) under anoxic and reducing conditions (Moore and Luoma, 1990; Morse, 1994). When exposed to atmospheric conditions either naturally or via anthropogenic activities like mining, mineral processing, metallurgy and underground space developments (Tabelin and Igarashi, 2009; Tabelin et al., 2012a, 2012b; 2014a, 2014b; 2017a, 2017b; Tamoto et al., 2015; Tatsuha et al., 2012), however, it is readily oxidized by oxygen (O_2) and ferric ion (Fe^{3+}) in the presence of water or moisture (Eqs. (1)–(3)), a process that generates acid mine drainage (AMD) containing not only Fe and sulfur (S) but also heavy metals (e.g., copper (Cu), lead (Pb), zinc (Zn) and manganese (Mn)) and toxic metalloids (e.g., arsenic (As) and selenium (Se)) because natural pyrite tends to incorporate these elements into its crystal structure (Tabelin et al., 2012c, 2017c; 2017d).



Arsenopyrite (FeAsS) is another important sulfide mineral especially in mining areas because its dissolution releases As (Eqs. (4) and (5)), a strictly regulated substance due to its toxicity at high concentrations, and its potential to cause numerous diseases like hyperpigmentation, keratosis, anemia, neuropathy and several types of cancers even at minute amounts (Boddu et al., 2008; Mohan and Pittman, 2007).



Neutralization is the most commonly used technique to treat AMD from metal sulfide mines (Johnson and Hallberg, 2005). In this technique, the acidic pH of AMD is raised using basic materials such as limestone (CaCO_3) or lime (CaO) to precipitate most of the dissolved metals (e.g., Fe, Mn and aluminum (Al)) (Goetz and Riefler, 2014; Kefeni et al., 2015). Although simple and effective, this technique has several limitations: (1) continuous supplies of chemicals and energy are required for very long periods of time (ca. tens–hundreds of years) (Gazea et al., 1996), and (2) problems with the disposal of bulky and hazardous sludges (Kefeni et al., 2015).

Because of these limitations, alternative methods aimed to limit the formation of AMD by directly passivating sulfide

minerals with a protective layer in a process generally referred to as encapsulation have gained attention in recent years (Diao et al., 2013; Evangelou, 1995; Liu et al., 2017). One promising technique that falls under this category is carrier-microencapsulation (CME) because it could specifically target pyrite and arsenopyrite in complex systems (Jha et al., 2008, 2011; Park et al., 2018; Satur et al., 2007; Yuniati et al., 2015). The unique targeting ability of CME could be attributed to the decomposition of metal(loid)-organic complexes only on surfaces of minerals that dissolve electrochemically like pyrite and arsenopyrite (Crundwell, 1988; Rimstidt and Vaughan, 2003; Tabelin et al., 2017c, 2017d). In CME, redox-sensitive organic compounds (e.g., catechol, 1,2-dihydroxybenzene) are used to transform relatively insoluble metal(loid) ions (e.g., Si^{4+} or Ti^{4+}) into soluble metal(loid)-organic complexes (e.g., $[\text{Si}(\text{cat})_3]^{2-}$ and $[\text{Ti}(\text{cat})_3]^{2-}$), which are stable in solution but decompose selectively on the surfaces of pyrite and arsenopyrite. The decomposition of metal(loid)-organic complex frees the insoluble metal(loid) ion, which is rapidly precipitated to form stable metal(loid)-oxyhydroxide coatings.

Although previous CME studies on $[\text{Si}(\text{cat})_3]^{2-}$ and $[\text{Ti}(\text{cat})_3]^{2-}$ showed promising results, the use of these complexes have some drawbacks. Formation of $[\text{Si}(\text{cat})_3]^{2-}$ requires very high concentration of catechol (Fig. S1) (Bai et al., 2011; Öhman et al., 1991) while the decomposition of $[\text{Ti}(\text{cat})_3]^{2-}$ was sluggish and took at least 14 days of treatment to generate a coating thick enough to be effective (Park et al., 2018). The high stability of $[\text{Ti}(\text{cat})_3]^{2-}$ could explain why its decomposition kinetics was slow, which delayed the formation of Ti-oxide coating. To accelerate coating formation and improve the practicality of CME, a new metal-catecholate complex that is easily synthesized and more readily decomposed is required as an alternative.

Al-catecholate complexes are promising candidates because according to Xu (2013), the cross-linking strength between metal and catechol weakens as oxidation state of the metal atom decreases. In other words, Al^{3+} -catecholate complexes are theoretically easier to decompose oxidatively compared with Ti^{4+} -catecholate complex, which could accelerate coating formation. Furthermore, AMD contains Al^{3+} at relatively high concentrations (100–500 mg/L) (Poza et al., 2017; Wei et al., 2005), which makes it a good candidate material for the synthesis of Al-catecholate complexes. Although several studies on Al-catecholate complex formation, stability and speciation have been conducted (Cornard et al., 2010; Nurchi et al., 2009; Sikora and McBride, 1989), very few have been done to understand the electrochemical properties of these complexes. To date, only the study of Downard et al. (1996) on the voltammetry of Al-catecholate complexes in buffered solutions has been reported, so the electrochemical behaviors of these complexes remain unclear. Furthermore, the interactions between sulfides (e.g., pyrite and arsenopyrite) and Al-catecholate complexes have not yet been reported.

In this study, Al-catecholate complexes were synthesized and their redox properties as well as their ability to suppress the oxidations of pyrite and arsenopyrite were investigated. Specifically, this study aims to: (1) elucidate the electrochemical properties of

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