

STUDY OF SULFIDE MINERALS OXIDATION SUPPRESSION USING SILICON-CATECHOL COMPLEX

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論 文 内 容 の 要 旨

Acid mine drainage (AMD) resulting from the oxidation of pyrite and other sulfide minerals has caused significant environmental problems, including the acidification of rivers and streams, and the leaching of toxic metals. Once exposed to water and oxygen during mining and mineral processing, sulfide minerals become immediately susceptible to chemical and biochemical oxidation. Passivation of sulfide minerals involving formation of a surface barrier through formation of a coating layer is a potential way to control AMD generation.

In this study, the promising method of carrier microencapsulation (CME) using a silicon-catechol complex to suppress sulfide minerals oxidation was investigated. To reveal the mechanism and coating layer properties, dissolution tests, and morphological and electrochemical analyses were performed. In addition, samples were characterized by surface-sensitive X-ray photoelectron spectroscopy (XPS), atomic force microscopy (AFM), and dynamic force microscopy (DFM).

This thesis consists of six chapters. **Chapter 1** presents the background and objectives of this study. In addition, an overview of AMD and its prevention are presented to provide a basic understanding of pyrite oxidation suppression. Previous studies related to the present study are also discussed in this chapter.

In **Chapter 2**, pyrite oxidation suppression by CME with silicon (Si) and an organic carrier is presented. It was found that waste water collected from hydrothermal treatment (HT) of low-rank coal contains organic carbon compounds, such as phenol and catechol. The use of HT liquid (HTL) produced from low-rank coal as a carrier in CME was investigated. In dissolution tests for 51 days with pyrite and iron-oxidizing bacteria, treatment with a mixture of HTL and a silicon reagent (Si-HTL) lowered the ferric ion concentration and limited bacterial attachment compared with untreated pyrite. These results indicate that pyrite oxidation can be suppressed simply by pretreating with Si-HTL for 1 h. This might be caused by catechol present in the HTL. A mixture of catechol and a silicon reagent (Si-Cat) was also used, and the coatings obtained using Si-HTL and Si-Cat were compared. Based on dissolution tests, the Si-HTL coating layer showed better barrier properties than the Si-Cat coating layer. Microscopic observations showed a silica-rich deposit on the surface of the treated pyrite. Fourier-transform infrared (FTIR) spectroscopy indicated that a quinone compound adsorbed on the treated pyrite surface. These results showed that silica and quinone were both adsorbed on the pyrite surface in a silica-quinone layer, which can suppress pyrite oxidation.

In **Chapter 3**, three different electrochemical methods, namely, potential polarization, chronoamperometry, and electrochemical impedance spectroscopy (EIS), were used to investigate the

electrochemical behavior of pyrite oxidation suppression in the presence of a silicate coating. Anodic current peaks corresponding to oxidative decomposition of the Si–Cat complex on the pyrite surface were observed at 550 mV in the presence of Si–Cat, and at 690 mV in the presence of Si–HTL. The anodic currents of the treated pyrite samples (Si–Cat pyrite and Si–HTL pyrite) were lower than that of the untreated pyrite. Because the anodic current represents the oxidation rate, the lower anodic currents of the treated pyrite samples mean that the oxidation rates of these treated samples are lower than that of the untreated pyrite. The two flat semi-circular curves in the Nyquist plots showed that the total impedances of the treated pyrite samples increased. It is suggested that this is a silica–quinone coating, which can decrease the pyrite oxidation rate. It also showed that the coating resistance produced by Si–HTL treatment is higher than that produced by Si–Cat. Bode plots showed the shift of the phase angle of the time constant of Si–Cat-treated pyrite to lower frequencies, indicating electrolyte diffusion occurs through the coating layer. This indicates that the coating layer produced by Si–HTL treatment has better barrier properties than that produced by Si–Cat and confirms the results of the dissolution tests and chronoamperometric measurements.

In **Chapter 4**, a more systematic investigation of the silica–quinone coating layer is presented. Furthermore, the mechanism involved in this coating treatment is proposed. Pyrite treatment using Si–Cat under different conditions resulted in differences in the suppression of pyrite oxidation. The electrochemical behavior of treated pyrite samples showed that Si–Cat treatment for 6 h with initial pH 9.5 gave better barrier properties and a higher suppression effect than that of other treatment conditions. FTIR and XPS analyses demonstrated that the coating layers on the treated pyrite samples consisted of a network of Fe–O–Si and Si–O–Si units on the pyrite surface. The Si–O–C asymmetric stretching mode is also present in the FTIR spectra. These results confirm that silicate polymerization in the silica–quinone layer on the treated pyrite samples suppressed pyrite oxidation.

In **Chapter 5**, pyrite oxidation suppression by the Si–Cat complex was applied to other sulfide mineral samples: chalcopyrite (CuFeS_2), molybdenite (MoS_2), and arsenopyrite (FeAsS). Similar to pyrite, the results indicate the formation of a coating layer on the treated sulfide mineral samples that lowered the oxidation rate. The Si–Cat complex showed a better suppression effect for chalcopyrite than for molybdenite and arsenopyrite.

Chapter 6 summarizes the conclusions of this study and provides recommendations for further study.