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Author(s)	TUM, Sereyroith
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学位論文題名

Geochemical Iron Dynamics and Passive Treatment of Acid Mine Drainage with High Dissolved Iron Concentration for Sustainable Green Mining

(持続可能なグリーンマイニングに向けた高濃度溶存鉄含有酸性鉱山廃水の地球化学的な鉄ダイナミクスの理解とパッシブトリートメント)

Acid mine drainage generated from dissolved metals sulfide is one of the world's environmental risks once it exposes to the environment. Typical toxic elements such as As, Ni, Cu, Se, Pb, Cd, and Fe release from AMD highly impact human health and aquatic lives. Some AMD cases are naturally attenuated without human interventions. Unfortunately, human assistant for water treatment is essential if natural attenuation is inadequate. Passive treatment is the sustainable method for AMD treatment, but it is dependent on site-specifics. Passive treatment is applicable only if a detailed study of natural remediation in AMD contamination sites. Ferric iron minerals such as schwertmannite, commonly found in an acidic environment, have potential roles in natural attenuation for heavy metal scavenges. Regardless of the significant roles of schwertmannite in natural attenuation, the gaps of the schwertmannite formation based on the iron dynamics study applied to natural environments remain uncertain. Therefore, the two case studies of AMD contamination are selected to investigate in this research to cover the broad scope of the relationship of iron dynamics and natural attenuations in the implication for passive treatment.

Chapter 1 introduces the research introduction, background, and problem statements. The overall objective of this research is to elucidate the effects of natural attenuation occurrences for various sources of iron species in acid mine drainage in different climate conditions and its implication for passive treatment methods. The implementation to reach these goals, the aspects that mainly focus on are: (1) the characterization of the various AMDs' geochemical behaviors in different climate conditions that possibly control the natural attenuation mechanisms, (2) elucidating the impacts of iron species and its dynamics that may well influence to AMD attenuations, and (3) the advantage of natural attenuation mechanisms to apply to AMD passive treatment methods by using geochemical modellings.

Chapter 2 reviews the existing literature to deliver this research's background knowledge. The appraisals include the roles of ferric iron minerals to remove toxic elements from AMD, the relationship of Fe dynamics, the formation of schwertmannite in the mine site, and the overview of AMD treatment methods.

Chapter 3 discusses the case study of Mondulkiri mine site, where the sources of AMD come from the discharge of tailing and AMD generation from the excavated area. Fe^{2+} is the major iron species released from AMD. The toxic elements which are more than the WHO regulation limit are As, Ni, Se, and Cu. Iron oxidizing bacteria in the tributary is one of the factors controlling the natural attenuation process. The rapid oxidation of Fe^{2+} to Fe^{3+} occurred under the biotic condition in the tributary. When pH increases in the rainy season, Fe^{3+} starts to precipitate in the tributary as schwertmannite, enabling As adsorption in the rainy season. Yet, Ni, Se, and Cu concentrations were diluted by rainwater. The natural attenuation successfully remediated the target contaminants to less than the WHO regulation limit in the rainy season. The tributary's pH decreases in the dry season led to the dissolution of

schwertmannite, where As was also released back to the river. The seasonal dynamics in the study area caught great attention of the role of rainwater or dilution in natural attenuation. In the rainy season, rainwater elevates the pH in the tributary from 2.7 to 3.4 and dilutes the contaminant concentrations. The dilution is insufficient to dilute As concentration to less than the environmental regulation limit, but the rising pH in the tributary facilitates schwertmannite precipitation. Thus, As adsorption onto schwertmannite occurs.

Chapter 4 is the case study of Shojin mine, the abandoned mine site in a temperate climate zone in northern Japan. The sources of the river contaminant are different from the Mondulkiri mine. The drainage seepage from the waste dams leaks to the nearby natural rivers rich in Fe, As, Cd and Pb. At the monitoring point, the contaminants of the Shojin river decreased to below the standard environmental level. In contrast, Cd and Pb concentrations in Amemasu river remain slightly higher than the standard environmental level, even though the two rivers have the same contamination sources. The Fe^{3+} is the dominant species released from drainage water, which is different from the Mondulkiri mine. After AMD had mixed with the Shojin river's pH decreased from 6.9 to 3.1. The formation of schwertmannite can be found just after mixing, and all the contaminant targets are less than WHO guidelines. Arsenic concentration decreases due to its adsorption onto the schwertmannite surface. Nevertheless, the concentrations of Pb and Cd were decreased by dilution of the natural water in the river. In contrast, Amemasu river's pH dropped from 7.1 to 2.8 after mixing with the drainage water. There was no significant effect of the natural attenuation in Amemasu river as it is in Shojin river.

Chapter 5 provides a detailed study of the iron dynamics in the study areas. The dominant ferric iron mineral found in both study areas is schwertmannite, has a significant role in natural attenuation. Iron dynamics in AMD control the rate-limiting of schwertmannite formation. Fe^{2+} is commonly found as the rate-limiting species in AMD treatment, yet in the case of Mondulkiri mine is different. Fe^{2+} is not the rate-limiting parameter. The presence of iron-oxidizing bacteria boosts the oxidation rate of Fe^{2+} to Fe^{3+} . Fe^{3+} precipitation rate constant of the study areas was obtained from the experimental data is relatively quick in acidic conditions at pH 3 to 3.5.

Chapter 6 and 7 describe each case study of toxic elements removal methods. The Acid neutralization by dilution and the utilization of limestone in AMD by the geochemical model was conducted to compare the efficiency of passive treatment implications. The Mixing models of the study areas were implemented to understand the role of dilutions in AMD contamination. The model shows the dilution could elevate the pH in rivers water and dilute the concentrations of the toxic elements. In addition, the drainage water reaction with the limestone model could also indicate the required limestone that is necessary for acid neutralization. Both methods provided significant results to the study area; however, the dilution of neutral water is limited to Amemasu river.

Chapter 8 concludes the natural attenuations mechanisms in each case studies and its future implications for passive treatment. This study shows some essential parameters that should be considered for passive treatment design, including iron species, iron concentration, dilution sources, flowrate, and etc. Moreover, this study validated the existed geochemical models and databases from previous studies to the measurement data from the natural environment. These findings are essential for the AMD contaminated area for future treatment implications.