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学 位 論 文 題 名

Effects of surface soil layer systems to remediate weathered rocks at a former mine site

（鉱山跡地における風化岩石の環境修復のための表層土壌層設置の影響）

学位論文内容の要旨

Pollution due to the formation of acid mine drainage (AMD) in former sulfide mines is a well known problem all over the world. The acidification of soil and water due to the oxidation of pyrite would generate a significant release of toxic heavy metals (e.g., copper (Cu), lead (Pb), zinc (Zn), and others) that could be transported in the surrounding aquatic environment over long distances and cause a serious health problem for humans when consumed directly (e.g., drinking) or indirectly (e.g., cooking). To deal with this problem, several researches have been done over the last decades, which tackle this issue with active (e.g., neutralization of AMD) or passive (e.g., reactive permeable walls for AMD and constructed wetlands) approaches. However, most of these remediation technologies require excessive amounts of money and/or energy and need to be monitored over long periods of time. In this context, an interesting novel approach is the utilization of layer systems to prevent the leaching of heavy metals and improve the acidic conditions, which can bringing back vegetation to these areas. Therefore, in this research we investigated the leaching characteristics and mechanism in the release of heavy metals from surface weathered rocks and evaluated the effectiveness of different soil layer systems at a flat and sloping surfaces at a former mine. These results will be used to design a remediation method for surface weathered rocks at the former mine sites under different topographical settings to enhance acidic environmental conditions.

Chapter 1 introduces the background of acidic soil contamination due to mining activities and current remediation technologies as well as the introduction of the soil layer system concept for engineering purposes. The statement of the problem and objectives of the study are also stated to organize the research activities.

Chapter 2 deals with the characterization of the selected closed mine site where the field observations were done and samples were collected. A brief introduction of the closed mine site as well as some historical monitoring data are described. The mineralogical and chemical compositions of the surface rocks indicated that conditions were acidic and trace amounts of sulfide minerals were found at the rocks that would continue to release heavy metals. In addition, the performance of candidate materials to be used in the layer systems was evaluated, which resulted in the selection of a mixture of $CaCO_3$ and surface rocks as the material to be used in the neutralization layer, clay collected near the site as the material to be used in the low-permeable layer, and silty soil collected from a nearby rice field as the material to be used in the vegetation layer.

In chapter 3, laboratory column experiments were carried out to elucidate the effects of different bedrock thicknesses on the leaching of heavy metals and major ions from the surface weathered rocks. The results showed that low pH and oxidating conditions were observed over a long period regardless of the thickness of the rock layer. However, a reduction in pH and an increase of leaching of heavy metals were observed with an increase in the thickness of the layer, except for Pb that did not follow this general behavior.

In chapter 4, we constructed three layer systems at a flat surface at the closed mine site by consider-

ing the characteristics of the site, the materials selected for the layers and the release of heavy metals from the rocks. The porewater chemistry, volumetric water content, and temperature in the layers were monitored over 20 months. When only a weathered rock layer was present, the pH values were acidic and the heavy metals were easily released into the porewater, but did not present any significant difference with regard to depth and season. However, when a neutralization layer was constructed on top of the weathered rock layer, the pH values were around the circumneutral region and concentration of heavy metals was drastically reduced. Moreover, when a low-permeable layer was included on top of the two layers described above, not only the pH remained in the circumneutral region and concentration of heavy metal were dramatically reduced, but also a reduction in the infiltration velocity of water though the layer was achieved. Thus, the final three soil layer system is recommended for the remediation of the flat surface.

In chapter 5, a similar evaluation as that in chapter 4 was done, when the topographical conditions were different. In this part, three layer systems were constructed on a sloping surface at the former mine to evaluate them. When only a weathered rock layer was used, low pHs and high concentrations of heavy metals were found, irrespectively of depth and season. Nevertheless, when a vegetation layer and soil cement layer (constructed to maintain the slope) were constructed on top of the weathered rock layer, pH values were around the circumneutral region and heavy metals were almost removed from the porewater. While the grown of vegetation in the layer (observed after the first winter season) at the surface prevented a rapid infiltration of water at the shallowest depths, the introduction of a low-permeable layer consisting of geomembrane underneath the vegetation and soil cement layers was effective in reducing the percolation. This last soil system appeared to be the most promising to remediate the acidic conditions and reduce rapid water infiltration. Also, a comparison between the results obtained for the weathered rock layer at the flat and sloping surface was done. A decrease in pH and an increase in the leaching of heavy metals for the sloping surface were observed compared to those for the flat surface, which suggests that an enhance of the acidification and heavy metal load in the porewater can be attributed to a longer interaction between rock and water due to the topographical condition.

Finally, a comparison of heavy metal leaching from the column experiments and that from field observations was discussed in chapter 6. An increase in the heavy metal release with a thicker rock layer was confirmed except for Pb. The main mechanisms of heavy metal leaching were found to be the two: the easy dissolution of labile phases and the ongoing oxidation of sulfide minerals. Also, the formation of anglesite ($PbSO_4$) as a secondary mineral was found to be the controlling phase in the dissolution of Pb from the rocks.

Chapter 7 provides the conclusions of the research as well as a tentative remediation scheme for the surface weathered rocks. This scheme combines a layer system for flat surfaces (neutralization and low-permeable layers) and a layer system for sloping surfaces (vegetation, soil cement, and low-permeable layers) to prevent rapid infiltration of water through the rock layer and to mitigate the acidic conditions of the weathered rocks, which would reduce the formation of AMD and improve the conditions for vegetation.

学位論文審査の要旨

主　査	教　授	五十嵐　敏　文
副　査	教　授	廣　吉　直　樹
副　査	教　授	高　橋　正　宏
副　査	准教授	原　田　周　作

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旧硫黄鉱山などにおける酸性坑廃水 (AMD) の生成にともなう汚染は世界中で知られている鉱業活動にかかわる環境問題である。黄鉄鉱のような硫化物の酸化による土壌・地下水の酸性化は，銅，鉛，亜鉛のような有害金属を溶出させ，周辺の水環境の悪化を招き，直接的あるいは間接的な摂取を通して人へ健康被害をもたらす懸念がある。この問題に対処するために，ここ数十年にわたり,AMD の中和のようなアクティブな処理や反応性透過壁や人工湿地によるパッシブな処理が行われてきた。しかし，これらの環境修復対策は長期にわたるコストやエネルギーが必要となる。この点で，重金属の溶出や酸性化を防止し，表層の植生を回復するために新たな表層土壌層を設置することは，有効な対策の一つとなる。本研究では，表層が風化した鉱山跡地において，そこから溶出する重金属の挙動を把握するとともに，多層からなる表層土壌層を平地と斜面に設置し，その有効性を評価した。本成果は，酸性環境にある鉱山跡地の環境修復対策を構築する上で有用であると考えられる。

第 1 章は鉱山活動にかかわる酸性土壌汚染の背景と従来型の環境修復対策を調査した上で，新たな表層土壌層設置の概念について検討するとともに，本研究の目的および構成について記述した。

第 2 章では，現地調査を実施した鉱山跡地の概要を記述した。表層風化岩石の化学的・鉱物学的特徴から，硫化鉱物が残存し，酸性環境のもと重金属の溶出が継続することが示された。また，表層に設置する中和層や難透水層に使用する材料の評価を行った。その結果，中和層には風化岩石と炭酸カルシウムとの混合土，難透水層には周辺で採取した粘土あるいは遮水シートを用いることとした。

第 3 章では，鉱山跡地で採取した風化岩石を異なる厚さで充填し，定期的に上部より脱イオン水を添加するという不飽和カラム試験を実施し，風化岩石からの重金属や主要成分の溶出挙動を明らかにした。その結果，溶出水の pH は，試験期間中低く，また重金属の溶出も継続して起こることがわかった。ただし，重金属の中で鉛の挙動は，充填厚さにあまり依存せず，銅や亜鉛の挙動と異なることが明らかになった。

第 4 章では，平地での現地試験方法とその結果を記述した。平地では，風化岩石だけからなるケース，風化岩石の上部に中和層を設けたケース，中和層の上にさらに粘土からなる難透水層を設けた 3 層からなるケースの 3 ケースの試験を実施した。各ケースに対して，特定深度に設置したポーラスカップから間隙水を定期的に採取した。また，温度・水分量・電気伝導度測定用センサーを特定深

度に設置し，連続的なモニタリングを実施した。その結果，風化岩石だけからなるケースは深度や季節にかかわらず,pH が低く，また重金属濃度も高かった。一方，中和層を設置したケースでは,pH が中性付近にまで上昇し，重金属濃度も激減した。さらに，難透水層を設置したケースでは，降雨にともなう水分量の変動が小さくなり降雨浸透量が減少した。このことから，難透水層と中和層を含む層構造が推奨された。

第 5 章では，斜面部での現地試験の方法とその結果を記述した。平地での試験と同様に，風化岩石だけからなるケース，風化岩石の上部に中和層を設けたケース，中和層の下部にさらに難透水層を設けた 3 層からなるケースの 3 ケースの試験を実施した。なお，斜面では粘土の敷設は困難であったため，難透水層として遮水シートを敷設し，中和層としてソイルセメントを吹き付けた。また，ソイルセメントの上部には畑地土を盛り付けた。風化岩石だけのケースは，深度や季節にかかわらず,pH が低く，また重金属濃度も高かった。一方，ソイルセメントを吹き付けたケースでは,pH が中性付近にまで上昇し重金属濃度は激減した。また，ソイルセメントと遮水シートによって降雨浸透量の抑制が認められた。平地における結果と比較すると，斜面における間隙水中の重金属濃度は高かった。このことは，表面流出量の相違に依存すると考えられる。

第 6 章では，室内カラム試験と現地試験結果とを比較した。銅や亜鉛では，充填厚とともに，あるいは現地での採水深度とともに，濃度が増加した。一方，鉛濃度は，充填厚や採水深度にかかわらずおおむね一定となった。地球化学解析によると，鉛はアングレサイト (硫酸鉛) の飽和指数がおおみねゼロとなった。このことは，鉛に対してはアングレサイトが濃度を制限していることを示す。

第 7 章では，以上の結果をまとめるとともに，難透水層と中和層，さらには植生層からなる構造が鉱山跡地では環境修復に有効となることを提案した。

以上を要するに，著者は，鉱山跡地の酸性土壌の環境修復および坑廃水の水量・水質改善のための合理的な方法として，中和層や難透水層からなる表層土壌層を設置することが有効であることを示しており，環境・資源工学の発展に寄与するところ大なるものがある。よって著者は，北海道大学博士 (工学) の学位を授与される資格があるものと認める。