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Title	On-site synthesis of schwertmannite and its application for arsenic immobilization at copper mines [an abstract of dissertation and a summary of dissertation review]
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Degree Grantor	北海道大学
Degree Name	博士(工学)
Dissertation Number	甲第12033号
Issue Date	2015-09-25
Doc URL	https://hdl.handle.net/2115/59997
Rights(URL)	https://creativecommons.org/licenses/by-nc-sa/2.1/jp/
Type	doctoral thesis
File Information	Houngaloune_Sengpasith_abstract.pdf, 論文内容の要旨

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学 位 論 文 内 容 の 要 旨
DISSERTATION ABSTRACT

博士の専攻分野の名称　博士（工学）　氏名　Houngaloune Sengpasith

学 位 論 文 題 名
Title of dissertation submitted for the degree

On-site synthesis of schwertmannite and its application for arsenic immobilization at copper mines
(銅鉱山におけるシュベルトマナイトのオンサイト合成とそのヒ素固定への利用)

Arsenic (As) is a very poisonous inorganic pollutant and a major toxicant at porphyry copper mines, the principal source of copper production worldwide. It is important to find a method to control or stabilize the toxic arsenic species that could be released from the large amounts of waste at copper mines. Among the current treatment processes for arsenic control of copper mine waste, retention of arsenic by schwertmannite (a ferric oxyhydroxy sulfate mineral) has attracted attention in recent years due to its strong binding affinity to toxic arsenic species. Schwertmannite based may also be cost-effective as it may be possible to synthesize schwertmannite from solutions generated in heap leach operations at copper mines. Leach solutions generally contain high concentrations of Fe^{3+} and SO_4^{2-}, the components of schwertmannite. In this study, on-site synthesis of schwertmannite at porphyry copper mines by neutralization is proposed. First, synthesis of schwertmannite from simulated copper heap leach solutions was investigated. The efficiency of arsenic removal by the synthesized schwertmannite and the stability of the arsenic-sorbed schwertmannite were then evaluated, and the application of schwertmannite in As control of copper mine waste was demonstrated.

In Chapter 1, the background and the objectives of the study are presented. The sources of arsenic contamination resulting from copper production are detailed. The various approaches to arsenic wastes control in mining and metallurgical operations are reviewed and schwertmannite was selected as the most promising compound for the control of toxic arsenic species in the large amounts of waste at copper mines. On-site synthesis of schwertmannite by increasing pH of the copper heap leach solutions is proposed and may be possible with pH 3-4. The synthesized schwertmannite can be expected to find application in arsenic immobilization of copper mine waste in tailings and spent ore of both dump and heap leach piles.

Chapter 2 details formation of schwertmannite in porphyry copper mines by investigating the effect of co-existing metal ions (Cu^{2+} and Fe^{2+}) and the reaction temperatures (25·C and 65·C) on the synthesis of schwertmannite by neutralization. It is shown that Cu^{2+} and Fe^{2+} play important roles in the schwertmannite synthesis at 65·C, however, Cu^{2+} and Fe^{2+} did not affect the synthesis at 25·C. It was observed that schwertmannite is formed at both 25·C and 65·C except, in solutions containing Fe^{2+}, where goethite was generated. With Fe^{2+} at 65·C, it was found that goethite is formed by transformation of schwertmannite, while Cu^{2+} has the ability to inhibit the transformation of schwertmannite to goethite in the presence of Fe^{2+}. Although it is possible to synthesize schwertmannite at both temperatures, their surface characteristics are different. The specific surface area of the schwertmannite synthesized at 65·C was much larger (147.4-176.9 $m^2\ g^{-1}$) than the specific surface area of the schwertmannite synthesized at 25·C (14.1-21.4 $m^2\ g^{-1}$), which this may affect their efficiency for arsenic removal.

In Chapter 3, arsenic sorption capacities by the synthesized schwertmannite were evaluated. The results indicate that As(V) in acidic solutions (pH 3-4) can be removed effectively by schwertmannite synthesized in the presence or absence of co-existing metal ions (Cu^{2+} and Fe^{2+}) at 65·C with the maximum sorption capacity of 94-133 mg g^{-1}. A lower As(V) sorption capacity is observed in product containing goethite synthesized in the presence of Fe^{2+} at 65·C; here, the maximum As(V) sorption capacity is 58 mg g^{-1}. The maximum As(V) sorption capacities by schwertmannite synthesized at 25·C are 17-23 mg g^{-1}, which are much lower than the maximum sorption capacities of the schwertmannite synthesized at 65·C. This should be taken into consideration for its application on mine sites.

In Chapter 4, the stability of As(V)-sorbed schwertmannite (Sch-As) under porphyry copper mining conditions

was studied by investigating the effect of Cu^{2+}, Fe^{2+}, pH, and ageing time on the stability of Sch-As. The results indicated that Cu^{2+} has no significant effect on the stability of Sch-As and that the As(V) incorporated into schwertmannite can retard or significantly inhibit the Fe^{2+}-catalyzed transformation of schwertmannite to goethite under acidic conditions (pH 3–4). The Sch-As aged at different pH ranges from 3 to 11 at 25·C exhibits no mineralogical phase changes even after ageing for 120-days; however the concentration of arsenic released from the solid phase appeared to be strongly pH-dependent also at ageing for 24 h. The release of As was almost negligible at pH 2 to 7, and a high release of As was observed at extremely acidic and alkaline conditions. This indicates that the release of As from Sch-As is controlled by environmental factors such as pH rather than time.

In Chapter 5, applications of the synthesized schwertmannite for arsenic immobilization in copper mine wastes were demonstrated. The study was divided into two parts. The first part was to demonstrate the application of schwertmannite for immobilization of arsenic in tailings resulting from copper flotation operations. The results of the experiments using a simulated tailings containing arsenopyrite (FeAsS) as arsenic-bearing minerals indicated that significant amount of arsenic were extracted to aqueous phase without the addition of schwertmannite due to the oxidation of arsenopyrite. The arsenic extraction deceased significantly when schwertmannite was added; suggesting that schwertmannite synthesized from copper heap leach solutions may be suitable for arsenic immobilization in copper flotation tailings. The second part of this chapter was focused on the possibility in applying the synthesized schwertmannite for concentrating arsenic from mine waters containing low concentrations of arsenic. The results indicated that the sorption-desorption process can be applied for this purpose: arsenic is removed from acidic solutions by sorption, and strip it back in alkaline solutions by desorption. By setting the sorption/desorption stripping ratio in the adequate value, arsenic can be concentrated into the alkaline stripping solutions. Higher concentrations of arsenic in the stripping solutions may be useful to form larger solid compounds, which have higher resistance against arsenic leaching.

In Chapter 6, the summary and conclusions of the study were presented.