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科学研究費助成事業　　研究成果報告書

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研究課題名（和文）Development of separation method for secondary copper and arsenic minerals using flotation

研究課題名（英文）Development of separation method for secondary copper and arsenic minerals using flotation

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研究成果の概要（和文）：本研究は、砒素を含む鉱物から銅を分離するプロセスを、浮遊法と過酸化水素を用いた酸化処理によって実証することに成功した。研究は、小規模な実験室実験から大規模なベンチ実験に至るまで、銅-砒素精鉱に対する酸化処理の基礎的な調査および実用的な実施を含んでいた。この研究により、最終銅精鉱中のヒ素濃度を、ヒ素を含む銅精鉱の処理基準値（0.2%）より低い値まで低減できることが実証された。さらに本研究は、ヒ素を含む銅精鉱から精製銅精鉱を生成する方法を提案している。

研究成果の学術的意義や社会的意義
This research developed a process to separate arsenic from copper concentrate using flotation and oxidation treatment, producing a clean copper concentrate that contains a minimal amount of arsenic. This will decrease arsenic emissions and minimize the pollution of the environment with arsenic.

研究成果の概要（英文）：This study successfully demonstrated the process of separating copper from minerals containing arsenic via flotation and oxidation treatment using hydrogen peroxide. The research included the basic investigation and practical implementation of the oxidation treatment on copper-arsenic concentrate, ranging from small-scale laboratory experiments to larger-scale bench experiments. This research demonstrated the ability to decrease the arsenic level in the final copper concentrate to a value lower than the threshold for processing copper concentrate with arsenic, which is set at 0.2%. Moreover, this research proposes a method for generating a purified copper concentrate from copper concentrate that contains arsenic.

研究分野：Mineral processing and flotation

キーワード：Flotation　Enargite　Chalcocite　Copper arsenic　Oxidation treatment　Hydrogen peroxide

１．研究開始当初の背景

Arsenic is an undesirable element in copper concentrate because of its toxicity and detrimental effects on human health and the environment, which diminishes the quality and value of the copper concentrate. Prior research conducted by Suyantara et al. (2020, Minerals Engineering, 152) has demonstrated that the application of oxidation treatment in selective flotation is a highly promising method for effectively separating arsenic-bearing copper minerals (enargite and tennantite) from primary copper sulfide minerals (chalcopyrite and bornite). Nevertheless, there is a scarcity of research on the specific separation of copper minerals containing arsenic and secondary copper sulfide minerals, such as chalcocite. Furthermore, there is a lack of detailed reports on the implementation of oxidation treatment for a copper-arsenic concentrate with higher complexity.

２．研究の目的

The aim of this research is to examine the effect of oxidation treatment using hydrogen peroxide (H_2O_2) on the floatability of chalcocite and enargite, both in the absence and presence of potassium amyl xanthate (KAX). This research examined the use of H_2O_2 treatment in the process of selectively separating chalcocite and enargite using flotation. In addition, this research investigates the impact of H_2O_2 treatment on the separation of arsenic and copper sulfide minerals in a complex copper-arsenic concentrate. This research aimed to identify the optimal conditions for producing a copper concentrate with minimal arsenic contamination and the greatest copper grade.

３．研究の方法

Pure chalcocite and enargite were crushed, hand-picked, dry-ground, and dry-sieved to obtain a particle size of 38–74 µm. Both chalcocite and enargite were mixed and then treated with KAX and various concentrations of H_2O_2 at pH 9 for 1 hour. The flotation test was carried out using a Partridge-Smith flotation glass cell and nitrogen gas. The froth was collected for 6 minutes, and the recovery was calculated based on the weight fraction of the dried froth.

A complex copper-arsenic concentrate was prepared with a pulp density of 37% (w/w). The suspension was agitated in Fahrenwald flotation equipment for 1 hour. Subsequently, the oxidation treatment was performed using 0.1 M H_2O_2 for 1 h. KAX was added, and the flotation test was performed. The froth concentrate was collected for 30 minutes and dried. The copper and arsenic recoveries were calculated and reported in this study.

４．研究成果

Figure 1 demonstrates the effect of H_2O_2 concentration on the floatability of chalcocite and enargite. The recovery of chalcocite had a modest increase after the oxidation treatment using a 1 mM H_2O_2 aqueous solution. However, the recovery of enargite had a substantial rise from 17% to 77% after the oxidation treatment using a 1 mM aqueous solution. The findings suggest that using a lower concentration of H_2O_2 during oxidation treatment may increase the capacity of enargite to float and preferentially separate it from chalcocite. Nevertheless, the use of intense oxidation treatment might hinder the capacity of both minerals to float and decrease the degree of separation selectivity. Figure 1 shows that the separation of chalcocite and enargite was most effective, reaching 50%, following the oxidation treatment using a 1 mM H_2O_2 solution at pH 9.

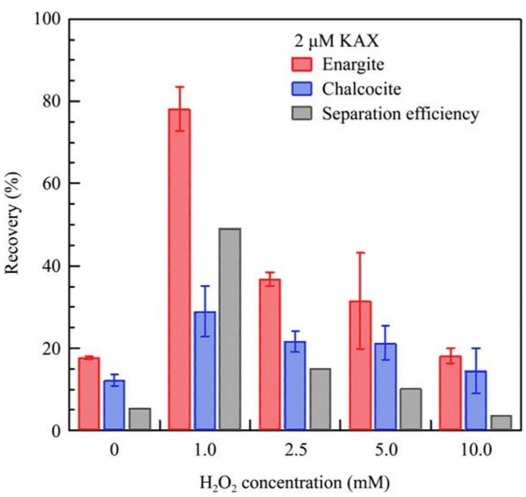

Figure 1. Effect of H_2O_2 treatment on the recovery of mixed chalcocite and enargite.

Figure 2 shows the effect of H_2O_2 oxidation treatment on the floatability of copper and arsenic sulfide minerals in a complex copper-arsenic concentrate. Figure 2 shows that the recoveries of total copper, arsenic, and non-arsenic copper after treatment with 60 g/t of KAX increased with increasing pH. The recovery of arsenic significantly improved by 29% after increasing the pH value from 9 to 10. The recovery of arsenic was 99% after treatment with 60 g/t of KAX at pH 10. The recovery of non-arsenic copper after increasing the pH value from 9 to 10 increased by 17%, which is significantly lower than the improvement in arsenic recovery. This is likely due to the more rapid adsorption of KAX onto the surface of arsenic-bearing copper minerals compared to that onto the surface of copper sulfide minerals at a higher pH value (alkaline condition).

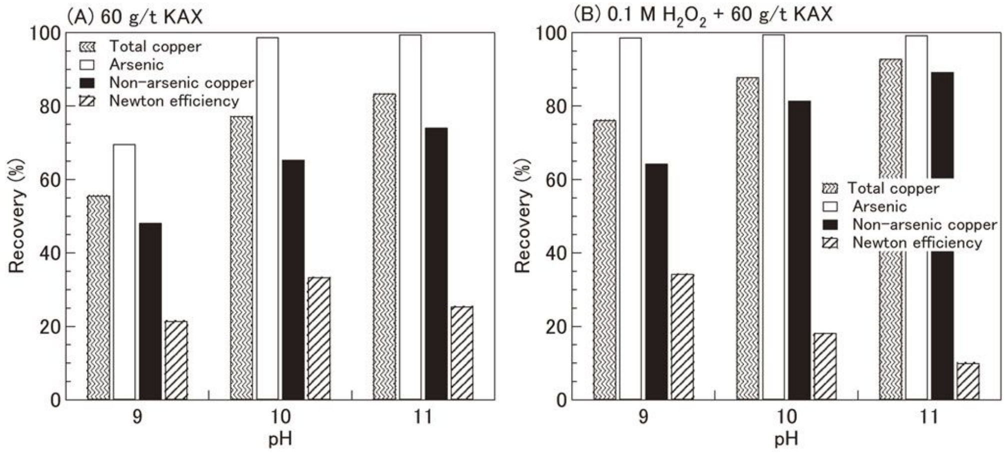

Figure 2. Effect of H_2O_2 oxidation treatment on the floatability of copper and arsenic sulfide minerals in a complex copper-arsenic concentrate at various pH conditions.

As shown in Figure 2, the optimum conditions (33%) for separating copper sulfide and arsenic-bearing copper minerals using 60 g/t of KAX was at pH 10. The Newton efficiency decreased at pH 11. This is due to a higher recovery of non-arsenic copper and the maximum recovery of arsenic. The arsenic level sharply decreased from 2.32% at pH 9 to 0.14% at pH 10 and 0.06% at pH 11 after treatment with 60 g/t of KAX and 30 min of flotation. These results indicate that producing a copper concentrate with a lower grade of arsenic, that is, lower than the penalty limit of 0.2%, is possible using the KAX treatment at the pH values of 10 and 11. These results demonstrate that KAX treatment under higher pH conditions can produce a "clean" copper concentrate with a lower arsenic grade. However, it should be noted that the grade of copper also decreased with increasing pH, that is, from 25% at pH 9 to 17% at pH 10 and 15% at pH 11.

Figure 2(B) presents the effect of oxidation treatment using a 0.1 M H_2O_2 aqueous solution and adding 60 g/t of KAX under various pH conditions on the recoveries of total copper, arsenic, and non-arsenic copper and Newton efficiency. The recovery of arsenic reached its maximum after 30 min of flotation at pH 9. Therefore, increasing the pH had no significant effect on the recovery of arsenic. In contrast, the recovery of non-arsenic copper increased with increasing pH. Accordingly, the Newton efficiency decreased with increasing pH. For instance, the Newton efficiencies were 18% and 10% at pH values of 10 and 11, respectively. Furthermore, the grade of copper decreased from 20% at pH 9 to 15% at pH 10 and 10% at pH 11 (Fig. 10(A)). Meanwhile, the grade of arsenic decreased slightly from 0.16% at pH 9 to 0.07% and 0.15% at pH values of 10 and 11, respectively. These results indicate that oxidation treatment using H_2O_2, followed by the addition of KAX at a higher pH was less effective for separating copper and arsenic-bearing copper minerals.

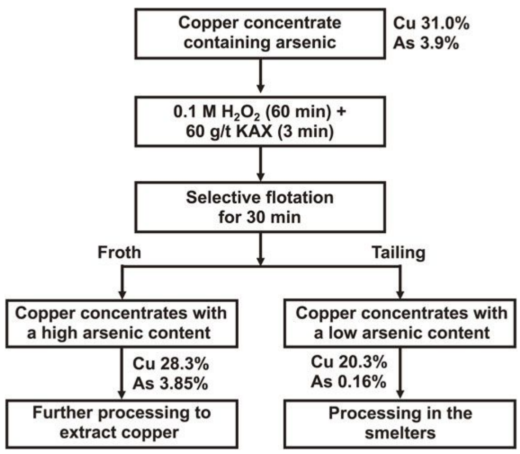

Figure 3. Proposed selective flotation process for the separation of arsenic and copper concentrate.

Based on these flotation results, we proposed a new process to separate copper and arsenic-bearing copper minerals via flotation, as shown in Figure 3. Selective flotation produces a "dirty" copper concentrate, which contains copper with high-grade arsenic, and a "clean" copper concentrate, which contains copper with low-grade arsenic in the tailing. The "dirty" copper concentrate can be further processed to extract copper from arsenic-bearing copper minerals, whereas the "clean" copper concentrate can be processed in smelters.

５．主な発表論文等

〔雑誌論文〕　計6件（うち査読付論文　6件／うち国際共著　6件／うちオープンアクセス　3件）

１．著者名 Suyantara Gde Pandhe Wisnu、Berdakh Daniyarov、Miki Hajime、Hirajima Tsuyoshi、Sasaki Keiko、Ochi Daishi、Aoki Yuji	４．巻 33
２．論文標題 Effect of hydrogen peroxide on selective flotation of chalcocite and enargite	５．発行年 2023年
３．雑誌名 International Journal of Mining Science and Technology	６．最初と最後の頁 703～716
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.ijmst.2023.01.002	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Berdakh Daniyarov、Miki Hajime、Hirajima Tsuyoshi、Sasaki Keiko、Ulmaszoda Akbarshokh、Nakao Ryota、Ochi Daishi、Aoki Yuji、Suyantara Gde Pandhe Wisnu	４．巻 431
２．論文標題 Effect of oxidation treatment on the selective separation of molybdenite from chalcocite using flotation	５．発行年 2024年
３．雑誌名 Powder Technology	６．最初と最後の頁 119078～119078
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.powtec.2023.119078	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Suyantara Gde Pandhe Wisnu、Miki Hajime、Ochi Daishi、Aoki Yuji、Ura Kumika、Berdakh Daniyarov、Ulmaszoda Akbarshokh、Dwitama Eska Putra、Sasaki Keiko、Hirajima Tsuyoshi	４．巻 34
２．論文標題 Sodium metabisulfite as a copper depressant in the selective flotation of copper-molybdenum concentrate using seawater	５．発行年 2023年
３．雑誌名 Advanced Powder Technology	６．最初と最後の頁 104258～104258
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.apt.2023.104258	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Suyantara Gde Pandhe Wisnu、Rizki Intan Nurul、Ulmaszoda Akbarshokh、Miki Hajime、Sasaki Keiko	４．巻 11
２．論文標題 Effect of goethite (α-FeOOH) nanoparticles on the surface properties and flotation behavior of chalcopyrite	５．発行年 2023年
３．雑誌名 Journal of Environmental Chemical Engineering	６．最初と最後の頁 110006～110006
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.jece.2023.110006	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Tanaka Yoshiyuki、Miki Hajime、Suyantara Gde Pandhe Wisnu、Aoki Yuji、Okamoto Hideyuki、Ura Kumika、Hirajima Tsuyoshi	４．巻 64
２．論文標題 Effect of pH and Precipitations on Copper?Molybdenum Rougher Flotation in Seawater	５．発行年 2023年
３．雑誌名 MATERIALS TRANSACTIONS	６．最初と最後の頁 1225～1231
掲載論文のDOI（デジタルオブジェクト識別子） 10.2320/matertrans.M-M2023805	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Suyantara Gde Pandhe Wisnu、Miki Hajime、Hirajima Tsuyoshi、Sasaki Keiko、Ochi Daishi、Aoki Yuji	４．巻 65
２．論文標題 Selective Flotation of Copper Concentrates Containing Arsenic Minerals Using Potassium Amyl Xanthate and Oxidation Treatment	５．発行年 2024年
３．雑誌名 MATERIALS TRANSACTIONS	６．最初と最後の頁 27～36
掲載論文のDOI（デジタルオブジェクト識別子） 10.2320/matertrans.M-M2023811	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

〔学会発表〕　計27件（うち招待講演　0件／うち国際学会　16件）

１．発表者名 Ryoji Ogi, Gde Pandhe Wisnu Suyantara, Hajime Miki, Tsuyoshi Hirajima, Keiko Sasaki, Yuji Aoki, Daishi Ochi
２．発表標題 Study on the Effect of Sodium Metabisulfite on Selective Flotation of Chalcopyrite and Molybdenite with Seawater
３．学会等名 International Symposium on Earth Science and Technology（国際学会）
４．発表年 2022年

１．発表者名 Daniyarov Berdakh, Gde Pandhe Wisnu Suyantara, Hajime Miki, Tsuyoshi Hirajima, Keiko Sasaki, Daishi Ochi, Yuji Aoki
２．発表標題 Separation of enargite and chalcocite with H2O2 oxidation treatment using flotation method
３．学会等名 International Symposium on Earth Science and Technology（国際学会）
４．発表年 2022年

１．発表者名
Daniyarov Berdakh, Gde Pandhe Wisnu Suyantara, Hajime Miki, Tsuyoshi Hirajima, Keiko Sasaki, Daishi Ochi, Yuji Aoki

２．発表標題
Separation of enargite and chalcocite with H2O2 oxidation treatment using flotation method

３．学会等名
The 16th International Symposium on East Asian Resources Recycling Technology（国際学会）

４．発表年
2022年

１．発表者名
Gde Pandhe Wisnu Suyantara, Tsuyoshi Hirajima, Hajime Miki, Keiko Sasaki, Yoshiyuki Tanaka, Yuji Aoki

２．発表標題
Effect of sodium sulphite on flotation behavior of bornite and tennantite

３．学会等名
International Mineral Processing Congress (IMPC) Asia Pacific（国際学会）

４．発表年
2022年

１．発表者名
Hajime Miki, Hiroki Fukuda, Berdakh Daniyarov, Gde Pandhe Wisnu Suyantara, Tsuyoshi Hirajima, Keiko Sasaki

２．発表標題
The estimation of microgram powder electrode system on sulfide mineral oxidation for mineral processing

３．学会等名
Copper 2022（国際学会）

４．発表年
2022年

１．発表者名
Gde Pandhe Wisnu Suyantara, Tsuyoshi Hirajima, Hajime Miki, Keiko Sasaki, Yoshiyuki Tanaka, Yuji Aoki

２．発表標題
Investigation of sodium sulfite as a selective depressant in the flotation of chalcopyrite and enargite

３．学会等名
Copper 2022（国際学会）

４．発表年
2022年

１．発表者名 Hajime Miki, Gde Pandhe Wisnu Suyantara, Tsuyoshi Hirajima, Keiko Sasaki, Yuji Aoki, Hiroki Kukuda
２．発表標題 Arsenic removal from arsenic containing copper concentrate with heat treatment and magnetic separation
３．学会等名 Copper 2022（国際学会）
４．発表年 2022年

１．発表者名 Ryoji Ogi, Gde Pandhe Wisnu Suyantara, Hajime Miki, Tsuyoshi Hirajima, Keiko Sasaki, Yuji Aoki, Daishi Ochi
２．発表標題 Study on the Effect of Sodium Metabisulfite on Selective Flotation of Chalcopyrite and Molybdenite with Seawater
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2022年

１．発表者名 Gde Pandhe Wisnu Suyantara, Tsuyoshi Hirajima, Hajime Miki, Keiko Sasaki, Yoshiyuki Tanaka, Eri Takida
２．発表標題 Oxidation treatment for separation of copper sulfide minerals and arsenic-bearing copper minerals using flotation
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2022年

１．発表者名 Daniyarov Berdakh, Hajime Miki, Gde Pandhe Wisnu Suyantara, Tsuyoshi Hirajima, Keiko Sasaki
２．発表標題 Selective flotation of Copper Mineral and Enargite with PAX and Hydrogen Peroxide
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2022年

１．発表者名
Gde Pandhe Wisnu Suyantara, Hajime Miki, Tsuyoshi Hirajima, Keiko Sasaki, Daishi Ochi, Yuji Aoki

２．発表標題
Investigation of sodium sulfite as a depressant for selective flotation of copper sulfide minerals from arsenic-bearing copper mineral

３．学会等名
The Mining and Materials Processing Institute of Japan Conference

４．発表年
2023年

１．発表者名
Rihan Efendi, Farahat Mohsen, Hajime Miki, Gde Pandhe Wisnu Suyantara, Keiko Sasaki

２．発表標題
Selective Flotation of Fluorite from Calcite using Sodium Humate as a Depressant

３．学会等名
The Mining and Materials Processing Institute of Japan Conference

４．発表年
2023年

１．発表者名
Eska Putra Dwitama, Akbarshokh Ulmaszoda, Gde Pandhe Wisnu Suyantara, Keiko Sasaki, Hajime Miki

２．発表標題
Evaluation of the Effect of Nanoparticles on the Flotation of Enargite

３．学会等名
International Symposium on Earth Science and Technology（国際学会）

４．発表年
2023年

１．発表者名
Akbarshokh Ulmaszoda, Gde Pandhe Wisnu Suyantara, Intan Nurul Rizki, Keiko Sasaki and Hajime Miki

２．発表標題
Investigation of the Effect of Goethite Nananoparticles on Chalcopyrite Floatability

３．学会等名
International Symposium on Earth Science and Technology（国際学会）

４．発表年
2023年

１．発表者名 Ryota Nakao, Hajime Miki, Eska Putra Dwitama, Keiko Sasaki, Daishi Ochi, Yuji Aoki, Gde Pandhe Wisnu Suyantara
２．発表標題 The Effect Of Various Reagents On Selective Flotation Of Molybdenite And Chalcocite
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Kazuto Satomura, Gde Pandhe Wisnu Suyantara, Hajime Miki, Keiko Sasaki
２．発表標題 Study on the Effect of Iron Oxide-Hydroxide Nanoparticles on Mixed Flotation of Bornite and Molybdenite
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Hajime Miki, Gde Pandhe Wisnu Suyantara, Keiko Sasaki
２．発表標題 Arsenic removal with heat treatment - magnetic separation from dirty copper conc.
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Hajime Miki, Tsuyoshi Hirajima, Gde Pandhe Wisnu Suyantara, Keiko Sasaki
２．発表標題 Evaluation of micro powder electrode on sulfide mineral leaching for hydrometallurgical process
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Eska Putra Dwitama, Akbarshokh Ulmaszoda, Gde Pandhe Wisnu Suyantara, Keiko Sasaki, Hajime Miki
２．発表標題 Preliminary Evaluation of the Impact of Nanoparticles on Enargite Flotation
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Gde Pandhe Wisnu Suyantara, Hajime Miki, Keiko Sasaki, Yuji Aoki, Daishi Ochi, Tsuyoshi Hirajima
２．発表標題 Utilizing Oxidation Treatment for Arsenic Mineral Removal in Producing a Cleaner Copper Concentrate using Flotation
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Akbarshokh Ulmaszoda, Gde Pandhe Wisnu Suyantara, Intan Nurul Rizki, Keiko Sasaki, Hajime Miki
２．発表標題 Investigation of Chalcopyrite Floatability in the Presence of Goethite Nanoparticles
３．学会等名 International Symposium on Metallurgical and Materials Engineering- The 18th International Conference on Quality in Research（国際学会）
４．発表年 2023年

１．発表者名 Hajime Miki, Gde Pandhe Wisnu Suyantara, Ryota Nakao, Kazuto Satomura, Keiko Sasaki, Daishi Ochi, Yuji Aoki
２．発表標題 The effect and mechanism of depressant on selective flotation of copper and molybdenum minerals
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2024年

１．発表者名 Gde Pandhe Wisnu Suyantara, Daniyarov Berdakh, Hajime Miki, Tsuyoshi Hirajima, Keiko Sasaki, Akbarshokh Ulmaszoda, Ryota Nakao, Daishi Ochi, Yuji Aoki
２．発表標題 Selective separation of molybdenite from chalcocite using flotation under oxidizing condition
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2024年

１．発表者名 Gde Pandhe Wisnu Suyantara, Akbarshokh Ulmaszoda, Hajime Miki, Keiko Sasaki, Taisuke Sakakibara, Hisatoshi Furuya, Satoru Okubo
２．発表標題 Study of pyrite depression for producing a high-grade copper concentrate from a complex copper ore
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2023年

１．発表者名 Ryota Nakao, Gde Pandhe Wisnu Suyantara, Hajime Miki, Keiko Sasaki, Yuji Aoki, Daishi Ochi, Satoru Yanagisawa, Eri Takida, Kumika Ura
２．発表標題 Effects of Sodium Metabisulfite and various collectors on selective flotation of Chalcocite and Molybdenite
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2023年

１．発表者名 Kazuto Satomura, Gde Pandhe Wisnu Suyantara, Hajime Miki, Keiko Sasaki
２．発表標題 Study on the Effect of Iron Oxide Nanoparticles on the Mixed Flotation Sorting of Mottled Copper Ore and Brilliant Lead Ore
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2023年

１．発表者名 Akbarshokh Ulmaszoda, Gde Pandhe Wisnu Suyantara, Hajime Miki, Daishi Ochi, Yuji Aoki, Kumika Ura, Daniyarov Berdakh, Eska Putra Dwitama, Keiko Sasaki, Tsuyoshi Hirajima
２．発表標題 Selective flotation of copper-molybdenum concentrate in seawater using sodium metabisulfite as a depressant for copper
３．学会等名 The Mining and Materials Processing Institute of Japan Conference
４．発表年 2023年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

資源処理・環境修復工学研究室 https://process.mine.kyushu-u.ac.jp/

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関