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Mineralogical characterization of the flotation products using automated mineral liberation analysis at the Erdenetiin Ovoo Cu-Mo porphyry deposit, Mongolia

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ABSTRACT

The flotation process is used to extract copper-molybdenum sulfide minerals from ore. The selection of the flotation technology scheme largely depends on factors such as the composition of sulfide ore in the ore body, grain size, and characteristics of the ore mineral association. The chemical and mineralogical analysis of flotation products was collected from the Erdenetiin Ovoo Cu-Mo porphyry deposit. The deposit is the largest porphyry copper-molybdenum deposit in Mongolia. The aim of this study was to demonstrate the occurrence mechanism of copper minerals in flotation tailing using the fully automated Tescan Integrated Mineral Analyzer. The chemical analysis of the flotation products (feed, concentrates and tailings) sample was conducted by X-ray fluorescence, and the mineralogical composition of the flotation feed sample was characterized using X-ray diffraction. The copper content of the flotation tailing was 0.024%. Mineralogical characterization results showed that almost all copper minerals occurred within coarse gangue particles, the primary and secondary copper minerals were accumulated in the size fractions less than 150 µm and 13.5 µm, respectively. The finest grain size distribution was observed in secondary copper particles of size -19 µm. Chalcopyrite was the main copper-bearing mineral, and it was closely associated with K-feldspar and silicate in the flotation tailings. The flotation tailing sample still contained 24.1 wt% liberated primary copper (chalcopyrite) and 24.13 wt% secondary copper due to their extremely fine grain size particle. The mineral map derived from Tescan Integrated Mineral Analyzer analysis revealed that copper minerals mainly occurred as finely disseminated and fully enclosed structures within gangue minerals.

Keywords: TIMA, phase boundary, association

INTRODUCTION

Flotation is a complex physicochemical process that is the most commonly used in mineral processing technology (Habashi, 2010). The process mineralogy of the feed, concentrate, and tailings of the flotation process will

provide solutions to improve the performance of the circuit in the flotation unit and suggest corrections to the plant flowsheet. The need for these studies is particularly accurate in sulfide minerals due to their complex mineralogical interactions (Chen et al., 2013; Qin et al., 2024).

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Copper sulfide ores, which contain primary and secondary copper sulfide minerals, are the main raw materials for copper extraction. About 80% of the copper mined worldwide is extracted from sulfide ores, especially in the form of chalcopyrite (Nyembwe et al., 2019; Han et al., 2017). The extraction of copper in sulfide ores from gangue minerals involves a cost-effective separation utilizing froth flotation, a process based on the physicochemical properties of mineral surfaces (Martínez-Gómez et al., 2016). Historically, flotation has been the preferred method for recovering copper from sulfide ores due to its notable flotation efficiency. However, several studies indicate that approximately 56% of the total copper within the ore is recovered from flotation methods (Antonijević et al., 2008; Han et al., 2018). Flotation residues from this process still contain valuable copperbearing minerals such as chalcopyrite and bornite. Consequently, numerous studies have focused on the reprocessing of copper tailings, recognized as a promising, cost-efficient, and environmentally sustainable approach enhancing copper production (Pazhoohan et al., 2019; Alcalde et al., 2018).

Mineralogical characterization has become a fundamental analysis in understanding an ore deposit and has been found important in interpreting metallurgical response, flowsheet design, and process selection, control, and optimization (Alcalde et al., 2018). The importance of process mineralogical studies and their influence on the flotation test has been well documented in research (Can et al., 2013; Tungpalan et al., 2015; Jefferson et al., 2023; Rincon et al., 2019; Whiteman et al., 2016; McKay et al., 2007). Automated mineralogy instruments such as QEMSCAN and mineral liberation analysis (MLA) have proven to be extremely useful in the field of process mineralogy and mineral processing. These techniques can provide quantitative mineralogical information, such as mineral compositions, elemental abundance, particle size. mineral interlocking, associations, and liberation characteristics, which is of great benefit for process optimization and improvement (Zhang et al., 2019; Liu et al., 2018; Li et al., 2018; Parker et al., 2015).

Lotter et al. (2003) studied the mineralogical characterization of flotation feed, concentrate, tailings by using polarized microscopy, and other techniques such as QXRD, QEMSCAN, EMAP, and SEM-EDS. Tiu et al. (2022) investigated the effect of sphalerite chemistry with grain size and liberation study using QEMSCAN®. Celep et al. (2019) highlighted the characterization of an extremely complex arsenical silver ore using MLA and provided invaluable insights into their refractory behavior. Furthermore, several authors (Mafra et al., 2020; Berkh et al., 2019) have utilized MLA for characterizing copper mine tailings, focusing solely on identifying the mineral composition and abundance of copper within the tailings.

Lately, the TESCAN Integrated Mineral Analyzer (TIMA) has also progressively found its way into many geological research laboratories where they are now being widely used for gathering mineralogical and petrological data. The TIMA system is an automated scanning electron microscope that provides particleby-particle quantitative mineralogical data on complex inorganic samples (Hrstka et al., 2018). It operates similarly to other scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS) techniques such as QEMSCAN and MLA (Pirrie et al., 2004; Haberlah et al., 2011), combining ultra-fast data acquisition (up to 100,000 mineral grains analyzed per hour via the collection of ~600,000 X-ray counts per second) with expert software to provide quantitative mineral and textural data down to a single particle (Ward et al., 2018).

In the available literature, the integrated automated mineralogy TIMA system has been used to characterize the mineralogy of the Rare-Earth Elements (REE) ore from the deep drill cores (Liu et al., 2022), to determine minerals and for mineral mapping in crushed lithium ore (Rifai et al., 2022), and to study the characterization of Ni/Co laterite deposits (Licia et al., 2020). From practical experience, it is known that the most common sources of unfavorable metallurgical indices of produced concentrates can be found in the mineralogical properties of flotation tailing. There is no specific data relating to the mineralogical properties of

copper molybdenum ore flotation tailings using the TIMA method.

The flotation products characterized in the present study originated from the Erdenetiin Ovoo Co-Mo porphyry deposit, one of the largest copper-molybdenum deposits in Mongolia. The deposit is hosted by an intrusive complex in the Orkhon-Selenge Trough (Sarantsatsral et al., 2021). The deposit was first discovered in 1964 and started to be mined in 1978. The mining area is mainly into four deposits including Central, Northwest, Shand, and Oyut.

Kim et al. (2014) found that the copper-bearing minerals in primary and secondary sulfide zones vary as follows: chalcopyrite, enargite, and tennantite are dominants in the primary ore zone, while chalcocite, covellite, and bornite occur in the secondary sulfide zone in Erdenetiin Ovoo Cu-Mo porphyry deposit. The objective of this study is to determine the occurrence mechanism of primary and secondary copper minerals, and the possible causes of copper losses in flotation tailings from Erdenetiin Ovoo Cu-Mo porphyry deposit. For this purpose, the full suite of quantitative mineralogical information obtained from the TIMA was presented and discussed. Mineralogical studies have been carried out on the concentrate and tailings of the flotation test. The liberation of valuable minerals, distribution of minerals at flotation feed and tailings, particle size for the flotation tailings, amount of copper sulfide minerals presents in the concentrate, amount of minerals lost in the tailings, and interlocking and mineral association for valuable minerals, were determined. Furthermore, X-ray fluorescence (XRF) was used to analyze the elemental composition of flotation feed and products. The research results have the potential to further improve copper recovery and provide theoretical guidance for optimizing the flotation process.

MATERIALS AND METHODS

Materials

The ore samples used in this study were collected from the future mining operation level (905-815 m) from the Erdenetiin Ovoo Cu-Mo porphyry deposit. The ore samples were crushed for 20 min with a jaw crusher to achieve a particle size distribution with a content of 65.7 wt% at <75

µm. Subsequently, samples of 1 kg were taken for grinding and flotation tests. To investigate the effect of chemical and mineralogical properties of sulfide minerals on the flotation process, a sample was collected from the labscale flotation test.

On the other hand, the same operation level and pre-lab crushed ore samples were used and grinded to the desired mineral size classifications for further study purposes. Epoxy resin was used as the background for the standard backscattered electron (BSE) image calibration. Based on the acquired BSE image, the different mineral phases were distinguished by their BSE grey values, and four EDAX element silicon drift detectors were applied for mineral identification and recognition. The samples were mounted with epoxy resin and were then polished and coated with a layer of carbon under vacuum, about 10-20 nm. The resin blocks with a diameter of 25 mm were placed into the chamber of the TIMA system and carried out the mineralogical analysis. The same preparation is done for the flotation products and tailings according to the desired size classification.

Flotation tests

The roughing process is the most crucial stage of the entire flotation process. It has a direct effect on the subsequent processes and the quality of the final products (Lu et al., 2018). Experimental flotation processes were carried out using a flotation machine (FML 240) with a 3.0-L cell and an impeller diameter of 70 mm. Considering the currently running flotation

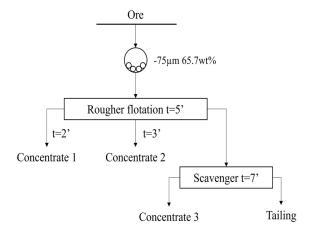


Fig. 1. Flowsheet of flotation experiment procedure

process at Erdenet Mining Corporation (EMC), lime (CaO) was selected as a pH modifier, commercial product BK-901 and isopropyl ethyl thionocarbamate (MF-03) were used as a collector, and methyl isobutyl carbinol (MIBC) was used as the frothers. The flotation tests were carried out using 6.0 g/t MF-03, 2.0 g/t BK-901, and 15.0 g/t MIBC. The flowsheet of the flotation experiments is shown in Fig. 1.

The ore samples (1.0 kg) were mixed with 600 mL of flotation water in ball mill feed. In each test, collectors and frothers were added to the flotation cells and mixed for approximately 1 minute. Slaked lime was then added to adjust the pulp pH to 10.5. In the next step, the air was blown into the airflow and the process of concentration was collected at 2 min, 3 min, and 7 min. Each product was filtered and dried at 100-105°C.

Characterization techniques

In this study, chemical and mineralogical analyses were performed at the Erdenet Mineral Research Institute of EMC. Elemental analysis of the flotation feed and products was conducted using X-ray fluorescence (PANalytical's MiniPal-4, Almelo, The Netherlands). The mineral compositions of ores were carried out using an X-ray diffractometer (D8 ENDEAVOR, Bruker, Germany).

Mineral liberation analysis of copper tailings can provide a wide range of mineralogical parameters, including mineral abundance, grain size distribution, mineral association, locking, middling, and, especially, the liberation characteristics of minerals (Tsend-Ayush, 2015). In this study, a mineralogical analysis was performed using a TIMA (Czech Republic) automated analyzer equipped with a BSE image and an energy-dispersive X-ray (EDX) combination (Hrstka et al., 2018). The TIMA system, which is available at Erdenet Mineral

Research Institute of EMC, is used for this study.

Behind the system, dot mapping method was carried out in each measurement. Depending on the BSE threshold, dot mapping applies a BSE grid with a particular resolution to either the entire sample or just a subset of its particles or grains. Dot mapping method provides an excellent compromise between the high-resolution mapping and the point spectrometry method in terms of speed versus textural detail.

RESULTS AND DISCUSSION

Chemical analysis

The chemical composition of the flotation feed and tailings samples from the production site were analyzed using XRF. The chemical composition of the collected samples for Cu, Mo, and Fe content is shown in Table 1.

As shown in Table 1, the content of copper, molybdenum, and iron in the feed is 0.248%, 0.019%, and 1.866%, respectively. The flotation products of Concentrate 1 (Con.1), Concentrate 2 (Con.2), and Concentrate 3 (Con.3) contained 6.02%, 1.113%, and 0.524% of copper, respectively. The copper content of the flotation tailing was 0.024%. The grade distribution of copper and another associated element (molybdenite and iron) in the feed, concentrates and final tailings of the flotation circuit is shown in Fig. 2. The results show that the copper content in the standard flotation test Con.1 is 6.02%. A notable 78% of the copper grade in the total concentrate was reached within the first 2 minutes of the Con.1 test, while Con.2 and Con.3 displayed a continuous decrease in the copper content within their concentrates. The content of copper, molybdenum, and iron in the tailings was significantly lower at 0.024%, 0.003%, and 1.85%, respectively. A grade of 1.85% Fe can be related to sulfide minerals containing Fe transferred to the tailing, such as

Table 1. Results of XRF analysis for samples from the flotation circuit

Sampling	Cu %	Mo %	Fe %
Rougher feed	0.248	0.019	1.866
Concentrate 1	6.020	0.246	12.05
Concentrate 2	1.113	0.071	5.850
Concentrate 3	0.524	0.030	4.940
Rougher tailing	0.024	0.003	1.850

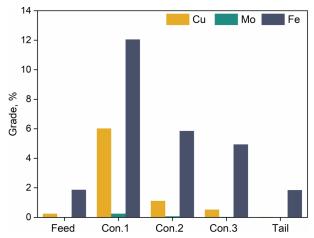


Fig. 2. Grade distribution of Cu, Fe and Mo metals in feed, concentrate, and tailings of flotation (Con.1: concentrate 1, Con.2: concentrate 2, Con.3: concentrate 3)

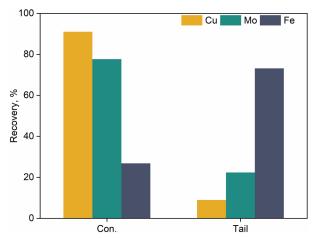


Fig. 3. Diagrams of Cu, Mo, and Fe recovery in final concentrate and tailings

Table 2. Cu, Fe, and Mo grade and recovery in final concentrate and tailings flotations

Sampling -	'	Recovery, %	
	Cu	Mo	Fe
Concentrate 1	83.31	68.04	20.71
Concentrate 2	5.69	7.25	3.71
Concentrate 3	2.06	2.35	2.41
Final concentrate	91.05	77.64	26.83
Tailing	8.95	22.36	73.17

chalcopyrite (CuFeS₂) and pyrite (FeS₂) which can be seen in Fig. 2.

A comparison of recoveries for Cu, Mo and Fe metals in the final concentrate and tailings of the rougher flotation results is shown in Fig. 3 and Table 2. The total recovery of Cu in the concentrate is 91.05% with the grade of copper only 7.66%. The recovery of Mo in the concentrate is 77.64% with a relatively high grade in the concentrate (at approximately 23%).

Mineralogical characterization

The ore mineral and rock-forming mineral contents of the flotation tailings are shown in Fig. 4A and Fig. 4B. The flotation feed contains 0.6 wt%, 0.5 wt%, and 0.2 wt% of the ore minerals such as chalcopyrite, chalcocite, and pyrite, respectively, as shown in Fig. 4A.

Fig. 4B shows the rock-forming minerals, and the results indicate that albite, quartz, muscovite, and orthoclase were the predominant minerals. Kim et al. (2013) investigated that pyrite are predominant ore mineral and that a significant amount of chalcopyrite and molybdenite were also observed in the Erdenetiin Ovoo Cu-Mo

porphyry deposit, but their contents in the tailings were so low that they could not be detected by XRD but only by more advanced technique. In this study, TIMA was employed to identify the mineral phase constituents of the flotation tailings. The result confirms that the tailings sample from the production site contained both primary and secondary copper sulfide minerals and pyrite was the dominant copper-bearing mineral phase.

Fig. 4C shows the amount of gangue minerals before and after flotation, while the amount of sulfide minerals decreased significantly after the flotation process. As presented in Fig. 4D, chalcopyrite and pyrite content in Con.1 of the flotation is 27.19 wt% and 15.42 wt%, respectively. However, according to the mineral mass distribution of the flotation tailings, chalcopyrite, and pyrite accounted for 0.86 wt% and 2.34 wt%, respectively, with the predominant presence of gangue minerals such as K-feldspar (orthoclase and microcline), quartz, and muscovite. This suggests that the majority of sulfide minerals in the flotation feed were successfully recovered in the concentrate products through froth flotation.

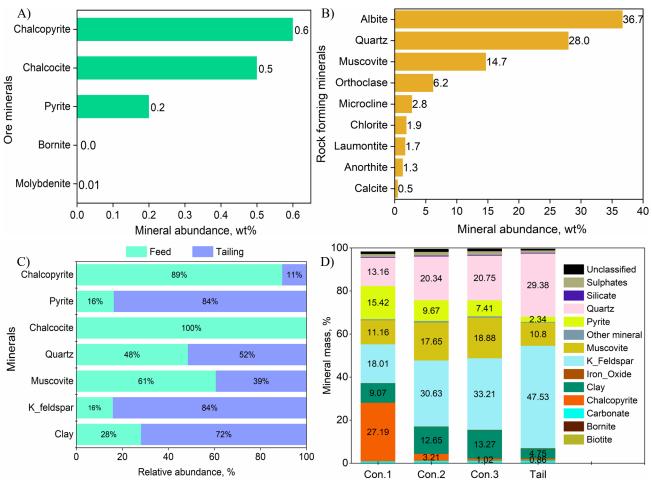


Fig. 4. Mineral constituents of the flotation feed and tailings samples: XRD mineral composition of A) ore minerals abundance B) rock-forming minerals abundance; C) relative mineral abundance determined by TIMA; D) results of the mineral mass distribution

Mineral grain size distribution

The knowledge of the flotation rate by mineral liberation allows the analysis and correlation between the original ore properties (original grain size distribution) with both the grinding product (size class distribution) and the flotation performance (grade/recovery by true flotation) (Vallejos et al., 2018). In this study, TIMA measurement provides a quantitative description of the grain size distribution in primary and secondary copper ore minerals. The grain size distribution of minerals in the flotation tailings is shown in Fig. 5. As can be seen in Fig. 5A, the primary and secondary copper minerals were accumulated in the size fractions less than 150 μm and 13.5 μm, respectively.

Cumulative passing minerals of 80 wt% (P_{80}) value helps determine if the milling process is achieving the desired particle size distribution for downstream processing. For instance, a lower P_{80} value indicates finer particles, which

may enhance the recovery of certain minerals but at a higher energy cost. The size corresponding to the P_{80} are pyrite (81.06 μ m), molybdenite (41.70 μ m), primary copper (34.65 μ m), and 10.76 μ m of secondary copper, respectively as shown in Fig. 5B. In the size class of -19 μ m, secondary copper exhibited the finest grain size distribution (Fig. 5C). The grain size distribution between primary and secondary copper was uniform, suggesting that most of them were locked together in the copper tailings.

The TIMA-derived image, sorted by the particle size of the flotation tailings, is shown in Fig. 5D. The result illustrates that primary copper (chalcopyrite) has occurred in a partially locked with coarse gangue particles such as K-feldspar, quartz, and muscovite. In some cases, the chalcopyrite is partially connected to the pyrite grains. Thus, further grinding is necessary to liberate the locked chalcopyrite, which leads to higher energy consumption.

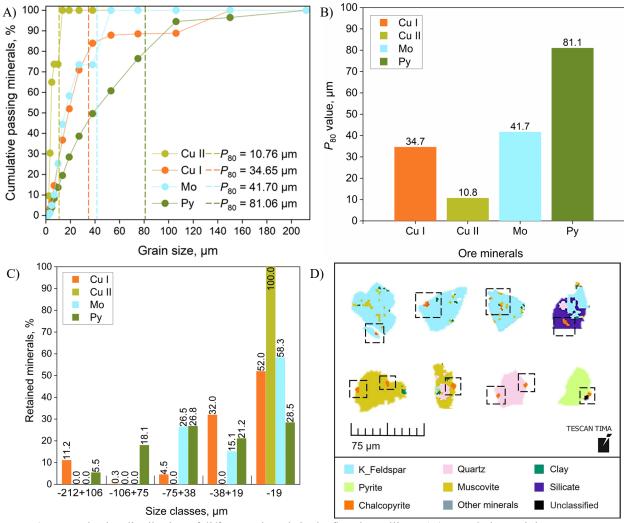


Fig. 5. Grain size distribution of different minerals in the flotation tailings: (A) cumulative weight percentage passing curve; (B) the grain size corresponding to the cumulative passing minerals of 80 wt% (P_{80}); (C) weight percentage retained in each class; (D) selected TIMA-derived image of chalcopyrite distribution

Mineral association

The association of copper minerals in the flotation tailings was determined using a TIMA analyzer. Fig. 6A shows the association profiles for sulfide minerals, including chalcopyrite, chalcocite, bornite, covellite, and pyrite. For sulfide minerals, the results of grain boundary detection suggest that there existed liberated grain boundaries that were not related to the gangue phase boundary. The free surfaces of chalcopyrite and bornite accounted for 45.07% and 59.31% of the total phase boundary. In addition, the length of the chalcopyrite phase boundary was associated with the K-feldspar, quartz, and muscovite by 19.59%, 12.75%, and 7.91%, respectively.

The distribution of chalcopyrite, which has been identified as the main copper-bearing mineral across different liberation classes, is presented in Fig. 6B. Silicate and K-feldspar were the predominant gangue minerals and were highly intergrown with unliberated chalcopyrite. The abundance of quartz in the unliberated chalcopyrite was significantly lower than that in the copper-free liberated fraction. Quartz, belonging to the common rock-forming minerals, is considered the dominant gangue mineral in copper sulfide tailings (Sracek et al., 2010). In terms of mineral content, clay, muscovite, and quartz accounted for 1.27 wt%, 1.26 wt%, and 1.17 wt%. Furthermore, K-feldspar was always the most abundant gangue mineral no matter the type of locking relationship. Compared with pyrite the occurrence mode of chalcopyrite was a bit more complex. The phenomenon that K-feldspar occurs as the most abundant silicate mineral with a comparably small amount of quartz is always found in tailings

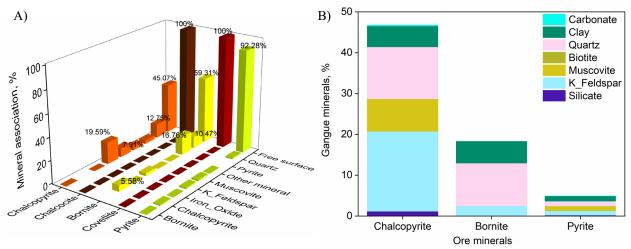


Fig. 6. Mineral association of the flotation tailings: A) copper minerals association with gangue minerals; B) copper sulfide minerals locking the relationship between major gangue minerals

Chalcopyrite Bornite K-Feldspar Muscovite Pyrite Molybdenite Quartz Clay Silicate 0.0580.840.001 0.059 0.04 3.21 5.59 0.08Chalcopyrite 0.014 0.02 0 0 0 0 0 0 Bornite 1.09 19.59 17.13 4.84 10.02 23.4 25.05 2.42 K-Feldspar 7.92 9.18 1.16 5.11 4.78 0 3.81 3.44 Muscovite 3.23 16.76 0.02 0.06 0 0.04 0.10 0.19 **Pyrite** 0 0 0 0 0.004 0.007 0.01 0 Molybdenite 12.75 10.47 6.58 6.66 1.17 30.74 4.45 4.48 Quartz 5.10 5.45 8.21 3.32 1.27 23.3 1.75 10.03 Clay 1.10 0 0.48 0.12 0 0.09 0.44 Silicate 45.08 59.31 73.04 69.26 92.29 29.57 80.35 61.17 49.38 Free surface

Table 3. Associations of minerals in flotation tailing

from alkalic Cu-porphyry mines such as Mount Polley (Kennedy et al., 2016). Therefore, the displacement of mineral contents derived from the results of mineral association can be used as a powerful parameter to evaluate the breakage characteristics (Parapari et al., 2022).

In Table 3 mineral associations of the investigated sample are presented. As it can be seen, copper minerals if they are not fully liberated, are associated mainly with clay, K-feldspar, quartz, and copper sulfides. It could be one of the critical reasons for the loss of copper in tailings.

Mineral liberation analysis

Fig. 7 presents the liberation characteristics of secondary and primary copper, molybdenite, and pyrite. As expected, the degree of liberation of copper minerals markedly became worse after the flotation process. Moreover, 85.1% chalcopyrite and 82.6% bornite were

accumulated in the <30% liberation classes, which suggests a lean intergrowth of copper-bearing minerals and gangue fractions, and the distribution of chalcocite in this fraction even reached 100%.

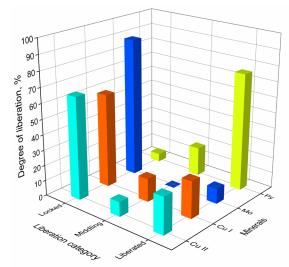


Fig. 7. Mineral liberation of secondary and primary copper, molybdenite, and pyrite in flotation tailing by liberation categories

	Č				
	Free surface %				
Mineral liberation	Primary copper (Cu I)	Secondary copper (Cu II)	Molybdenite (Mo)	Pyrite (Py)	
Locked <20	60.75	66.30	90.01	5.38	
Middling ≥20<80	15.15	9.57	0	18.79	
Liberated ≥80	24.10	24.13	9.99	75.83	
Total	100	100	100	100	

Table 4. The liberation of ore minerals in the flotation tailing

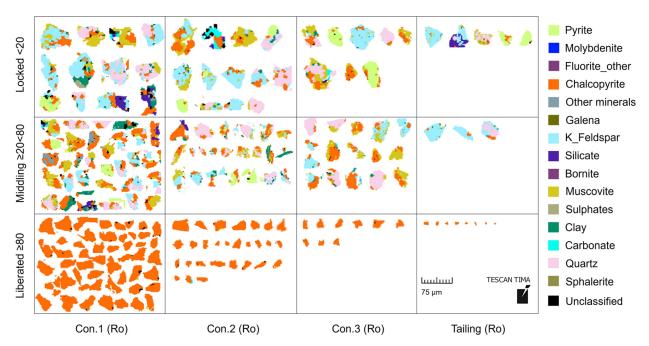


Fig. 8. Selected TIMA image of chalcopyrite in flotation Con.1, Con.2, Con.3 and tailings sorted by liberation categories

The selected TIMA image of the flotation products (Con.1, Con.2, Con.3 and tailings) from the production site, in which chalcopyrite is sorted by liberation categories, is presented in Fig. 8. The cross-sectional image illustrates that the chalcopyrite is finely dispersed in the flotation tailings and wrapped in gangue mineral particles. Although the tailings contained liberated chalcopyrite particles, accounted for 24.1%. The particle size of liberated chalcopyrite was less than 30 µm, whereas 52% had a particle size less than 20 μm. Zhang et al. (2021) found that liberated chalcopyrite with particle size less than 20 µm has a high probability to be lost in the tailings due to its ultra-fine grain size. As shown in Fig. 7. and Fig. 8, for primary and secondary copper, 60.8% and 66.3% were partially locked within gangue minerals such as pyrite, quartz, and K-feldspar, respectively, whereas for molybdenite, the value rises to 90.0% in the tailings. It is inferred from the results that copper minerals exhibited an extremely poor degree of liberation in the tailings, and molybdenite had the worst liberation degree as a valuable mineral. Due to either association with gangue minerals and ultrafine size are the main causes for the loss of copper-bearing and molybdenite minerals in the tailings.

The results of this study provide important insights into the ore mineralogy of the Erdenetiin Ovoo Cu-Mo porphyry deposit. The automated quantitative mineralogy using the TIMA has provided valuable information on the ore mineralogy of the Erdenetiin Ovoo Cu-Mo porphyry deposit. The results of this study can be used to guide further exploration and development of the deposit.

CONCLUSIONS

In this study, a comprehensive mineralogical characterization was carried out using fully automated TIMA to further clarify the occurrence mode of copper in flotation tailings from the Erdenetiin Ovoo Cu-Mo porphyry deposit. The mineral map of the TIMA image

confirmed that most of the primary copper mineral in the tailing was completely enclosed by the gangue minerals. The unrecovered copper in the tailings occurred in a few liberated ultrafine particles and interlocked structures; these structures were exceedingly dispersed and enclosed. The obtained results are summarized as follows:

- 1. The study revealed that chalcopyrite was the dominant valuable mineral, highly intergrown with K-feldspar and muscovite rather than quartz.
- 2. The phase boundary breakage of chalcopyrite-muscovite was relatively weaker than that of chalcopyrite-K-feldspar, whereas bornite tended to be more associated with the quartz and pyrite phase boundary.
- 3. The liberated chalcopyrite grains, accounting for 24.1wt%, remained unrecovered in the flotation tailings due to their extremely fine grain size of the former less than 150 μm.
- 4. Copper minerals predominantly occurred as fine-grained distribution within gangue minerals. The area percentages of primary and secondary copper minerals completely encapsulated by gangue minerals were 60.75% and 66.30%, respectively, whereas the proportion reached 90% for locked molybdenite. The degree of liberation of chalcopyrite in concentrates decreased, while the pyrite liberation increased in contrast.
- 5. The results of this study have significant implications for the recovery of residual copper from the flotation tailings.

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Statement of Author Contribution and Conflicts of Interest

B.T. and G.Ch. conceived and supervised the study. B.T., Ch.B., and Ts.Ts. developed the theoretical framework and performed the experiments. G.D. and N.A. aided in the analysis. D.B., S.G., and A.D. discussed the results and contributed to the final manuscript. The authors declare no conflicts of interest.

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