IJMMM
International Journal of
Minerals, Metallurgy and Materials
矿物冶金与材料学报（英文版）

Mineralogical characterization of copper sulfide tailings using automated mineral liberation analysis: A case study of the Chambishi Copper Mine tailings

Xiao-liang Zhang, Jue Kou, Chun-bao Sun, Rui-yang Zhang, Min Su, and Shuo-fu Li

Cite this article as:
Xiao-liang Zhang, Jue Kou, Chun-bao Sun, Rui-yang Zhang, Min Su, and Shuo-fu Li, Mineralogical characterization of copper sulfide tailings using automated mineral liberation analysis: A case study of the Chambishi Copper Mine tailings, *Int. J. Miner. Metall. Mater.*, 28(2021), No. 6, pp. 944-955. https://doi.org/10.1007/s12613-020-2093-1

International Journal of Minerals, Metallurgy and Materials
Volume 28, Number 6, June 2021, Page 944
https://doi.org/10.1007/s12613-020-2093-1

Mineralogical characterization of copper sulfide tailings using automated mineral liberation analysis: A case study of the Chambishi Copper Mine tailings

Xiao-liang Zhang[1)], *Jue Kou*[1)], *Chun-bao Sun*[1)], *Rui-yang Zhang*[1)], *Min Su*[2)], *and Shuo-fu Li*[1)]

1) School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China
2) Non-ferrous Corporation Africa Mining Public Limited Company, Kitwe 22592, Zambia
(Received: 18 February 2020; revised: 6 May 2020; accepted: 9 May 2020)

Abstract: As ore grades constantly decline, more copper tailings, which still contain a considerable amount of unrecovered copper, are expected to be produced as a byproduct of froth flotation. This research reveals the occurrence mechanism of copper minerals in typical copper sulfide tailings using quantitative mineral liberation analysis (MLA) integrated with scanning electron microscopy–energy dispersive spectroscopy (SEM–EDS). A comprehensive mineralogical characterization was carried out, and the results showed that almost all copper minerals were highly disseminated within coarse gangue particles, except for 9.2wt% chalcopyrite that occurred in the 160–180 μm size fraction. The predominant copper-bearing mineral was chalcopyrite, which was closely intergrown with orthoclase and muscovite rather than quartz. The flotation tailings sample still contained 3.28wt% liberated chalcopyrite and 3.13wt% liberated bornite because of their extremely fine granularity. The SEM–EDS analysis further demonstrated that copper minerals mainly occurred as fine dispersed and fully enclosed structures in gangue minerals. The information obtained from this research could offer useful references for recovering residual copper from flotation tailings.

Keywords: copper tailings; chalcopyrite; mineral liberation analysis; particle size; boundary breakage

1. Introduction

Copper sulfide ores, which contain primary and secondary copper sulfide minerals, are the chief raw materials for copper extraction. Approximately 70%–80% of the world's copper is extracted from sulfide ores, particularly as chalcopyrite [1–3]. The copper in sulfide ores can be economically separated from gangue by froth flotation, which is a separation process based on the physicochemical properties of the mineral surface [4–5]. Thus far, flotation has been the preferred method for recovering copper from sulfide ores because of the relatively good floatability. However, it has been reported that the total copper recovered from its ore by flotation is only about 56% [6–7]; a few valuable copper-bearing components such as chalcopyrite and bornite are still present in the flotation tailings. Thus, numerous studies have been focused on the reprocessing of copper tailings, which is considered a promising, economical, and environment-friendly approach for copper production [8–11].

With the dramatic increase in the global copper demand, a large volume of main tailings has been produced as a byproduct of flotation over the past few decades [12]. Sulfide tailings generated from the engineering process of copper mines are estimated to be more than 30 million tons all over the world, of which almost 90%–95% are produced from the flotation process [13–14]. Given that high-grade copper ores have been mined preferentially, the amount of copper tailings will continue to increase with declining ore grades. Considering this, it is increasingly important to improve the flotation performance and copper recovery efficiency of the current flotation circuit. Asghari *et al*. [15] explored the possible causes of copper loss to tailings and improved the performance of the Sarcheshmeh copper plant flotation circuit. Mackay *et al*. [16] studied the effects of particle size and superficial gas velocity on the dynamic froth stability of copper tailings. Zhu *et al*. [17] developed a novel collector (ZH-1, C3-5 carbon chain xanthate) to effectively improve the recovery of ultrafine copper oxide ore. Feng *et al*. [18] pointed out that a combined use of acidified water glass and locust bean gum could be applied for the effective separation of copper sulfides and multiple magnesium silicate minerals.

The global average grade of copper ore mined is 0.62%, and the average copper content in copper deposits is approx-

Corresponding authors: Chun-bao Sun E-mail: suncb@ustb.edu.cn; Jue Kou E-mail: koujue@ustb.edu.cn

Springer

imately 0.4wt% [19–20]. With the development of separation techniques, the copper content in the flotation tailings has fallen from 0.75wt% to 0.14wt% [21]. Lü *et al.* [22] found that ammonium humate was an effective regulator, through which the copper content of tailings was further lowered to 0.06wt%. The Chambishi Copper Mine is located in northwestern Zambia, belonging to the Central African Copperbelt. It is believed to be the world's largest sediment-hosted stratiform copper deposit [23–25]. It is a typical copper sulfide mine, and its copper reserve is estimated as 7 million tons, with an average copper grade of 2.07% [26]. The copper flotation tailings obtained from the mine have been reported to still contain 0.26wt% copper, which is larger than the economic grade (0.07%) for recovering single copper element [27–28]. Therefore, to improve the recovery of copper and maximize profits, it is essential to find the possible causes of copper losses to the flotation tailings.

The process mineralogical characterization of copper tailings, which involves the liberation and separation of the interested mineral, is essential to improve the performance of flotation circuits [29]. Mineral liberation analysis (MLA), an automated mineralogy characterization technique, has shown excellent application value in the fields of process mineralogy and mineral processing [30–31]. This technique can provide quantitative mineralogical information, such as mineral compositions, elemental occurrences, practical size, mineral locking, and liberation characteristics, which is of great benefit to process optimization and improvement [32–35]. For example, Hoang *et al.* [36] investigated the mineralogy of a carbonaceous sedimentary apatite ore using MLA and optimized the dominant hydrodynamic parameters of the flotation process. Moreover, a mineral liberation analyzer has been used to provide mineralogy parameters for rare-earth ores and improve the beneficiation process [37]. In one study, the range of mineralogical parameters obtained using the automated analysis technology provided strong support for the reprocessing of Zn tailings [38]. Furthermore, several authors [39–40] have also applied MLA in the characterization of copper mine tailings, but they only identified the mineral composition and abundance of copper tailings. Wang and Wang [41] studied the dissemination state of copper in an old mine tailing using X-ray diffraction (XRD) analysis, scanning electron microscopy (SEM), and MLA. They found that 31.11% Cu occurred as adsorbed particles or micro-particle inclusions in clay minerals, but the detailed mineral liberation and association information was not presented and analyzed. Naumov *et al.* [42] applied the characterization technique to study the mineralogy of selected samples taken during a plant survey for the Chelopech mine. The authors attributed the cause of copper losses to non-floatable chalcopyrite and particle locking.

The objective of this study is to determine the occurrence mechanism of copper minerals and the possible causes of copper losses in flotation tailings from the Chambishi Copper Mine. For this purpose, the full suite of quantitative mineralogical information obtained from MLA was presented and discussed. Moreover, SEM coupled with energy-dispersive X-ray spectroscopy (SEM–EDS) was used to analyze the powder sample and polished sample to further clarify the occurrence of copper in the tailings. The research results have the potential to further improve copper recovery and provide theoretical guidance for optimizing the flotation process.

2. Experimental

2.1. Materials

The ore samples used in this study were collected from the currently running processes at the Chambishi Copper Mine, located in the south-central part of Zambia. A plant survey of the flotation circuit was conducted twice to collect samples from determined sampling points. The interval time of each sampling was fixed at 30 min, and the sampling procedure lasted 5 h. The dry weights of the flotation feed and tailings samples were both approximately 6 kg. The obtained samples were further split for chemical assays and process mineralogical characterization. The product fineness for the flotation feed in an industrial flotation circuit was around 70wt% passing through 200 mesh. The main recoverable valuable element in the ore samples was copper. Its content was reduced to 0.26wt% after the flotation process, as determined by inductively coupled plasma–optical emission spectroscopy. In addition, the flotation tailings sample contained predominantly 56.60wt% SiO_2 along with 16.20wt% Al_2O_3 and 10.60wt% K_2O (Table 1).

Table 1. Main chemical components of the flotation feed and tailings samples obtained by XRF wt%

Sample	K_2O	CaO	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	Fe_2O_3	CuO
Flotation feed	10.20	4.28	0.25	6.12	17.90	49.30	0.18	2.17	5.97	2.04
Flotation tailings	10.60	4.66	0.31	4.66	16.20	56.60	0.21	0.43	4.30	0.35

2.2. Flotation tests

The roughing procedure is the most vital stage of the whole flotation process. It has a direct impact on the subsequent procedures and the quality of final products [43]. Thus, a single-stage roughing flotation process was designed for the bench-scale flotation tests to recover the residual copper in the flotation tailings. Flotation tailings with particle size larger than 0.1 mm was used as the ball mill feed, and the flotation tests were conducted using a 1.0 L XFD series mechanical agitation flotation machine. Considering the currently running flotation process at the Chambishi Copper Mine, lime (CaO) was selected as a pH modifier; the commercial product sodium isopentyl xanthate (SIX) was used as the copper collector, and 2# oil was used as the frother. All reagents used in the study were obtained from the Chambishi dressing plant to avoid unnecessary interference. The single-stage flotation tests were carried out using 600 g/t lime, 40 g/t SIX, and 25 g/t frother. The flowsheet of the lab-scale flotation experiments is shown in Fig. 1.

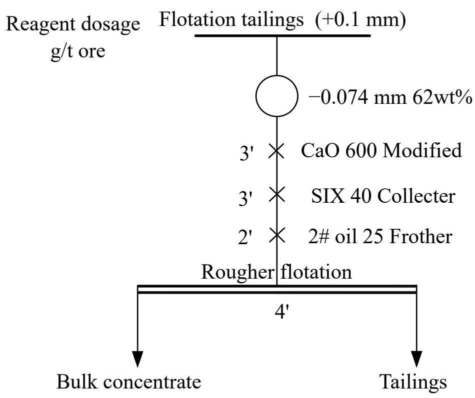

Fig. 1. Flowsheet of the lab-scale single-stage flotation tests.

2.3. Characterization techniques

Elemental analysis of the flotation feed and tailings samples was carried out using X-ray fluorescence (XRF) spectroscopy, and the copper content was accurately determined by inductively coupled plasma–optical emission spectroscopy after acid digestion. The mineral composition of ores was identified using a Rigaku Ultima IV X-ray diffractometer fitted with a Cu K_α radiation source (λ = 0.15406 nm, 40 kV, 40 mA). X-ray diffraction patterns were recorded from 10° to 90° (2θ angle) with a scanning speed of 20°/min. Moreover, SEM–EDS analysis was performed on the powder sample and polished sample using a scanning electron microscope (Zeiss, model EVO-18) coupled with an energy–dispersive spectrometer (Bruke XFlash Detector 5010) operated at 20 kV in a high vacuum.

Mineral liberation analyzer measurements of copper tailings can provide an extensive range of mineralogical parameters, including mineral abundance, grain size distribution, mineral association, locking, and, especially, the liberation characteristics of minerals [44]. In this study, a mineralogical analysis was performed using a mineral liberation analyzer from the Institute of Process Engineering, Chinese Academy of Sciences, which comprises an FEI Quanta 250 FEG scanning electron microscope equipped with an energy dispersive X-ray spectrometer (EDS) and the software package MLA suite 3.1.1.280 for data processing. The MLA measurement was carried out using the centroid mode to generate particle data. The calibration of the backscattered electron (BSE) greyscale with contrast and brightness was performed with gold reference. Epoxy resin was used as the background for the standard BSE image calibration, and gold as the upper limit (BSE grey value >250) [45–46]. Based on the acquired BSE image, the different mineral phases were distinguished by their BSE grey values and an EDAX Genesis EDS system was applied for mineral identification and recognition. The samples were mounted with epoxy resin and were then polished and coated with a layer of carbon, about 10–20 nm. The polished sections were observed and analyzed under the BSE mode with an acceleration voltage of 30 kV and 4 nm resolution.

3. Results and discussion

3.1. Copper element distribution

A sieve analysis experiment was conducted to determine the copper distribution in different particle-size fractions, and the results are shown in Fig. 2. The copper content was less than 0.75wt% in each particle-size fraction, and a significant negative correlation relationship existed between copper grade and particle size (Fig. 2(a)). Fig. 2(b) illustrates the copper distribution across various particle-size fractions. For the size fraction larger than 0.1 mm, the yield was 24.48%, but its copper amount accounted for 57.94wt% of the total copper. This implies that a significant amount of copper in the sample occurred as coarse mineral particles. The copper content in the finer size fraction less than 0.038 mm was only 0.1wt%, but the yield of this size fraction was high enough (44.55%) so that the amount of copper still accounted for 21.16wt% of the total copper. This is because fine particles are more likely to be liberated than coarse particles, but the finer copper-bearing particles always exhibit poor flotation performance [47]. The results of sieve analysis confirm that the unrecovered copper is mainly accumulated in the two size fractions: those larger than 0.1 mm and those less than 0.038 mm; the particle size needs to be further reduced to liberate the copper minerals.

Single-stage flotation tests were performed to study the effect of regrinding on the flotation performance of the flotation tailings sample with particle size larger than 0.1 mm. As listed in Table 2, the copper recovery of concentrate products was improved from 30.78% to 74.59%. Furthermore, the copper content decreased from 0.32wt% to 0.13wt%, which suggests that copper minerals remained locked in the coarse particles larger than 0.1 mm. The reground flotation tailings exhibited significant advantages regarding copper recovery over the unground tailings. The results confirm that further fine-grinding is beneficial to the recovery of residual copper in the flotation tailings.

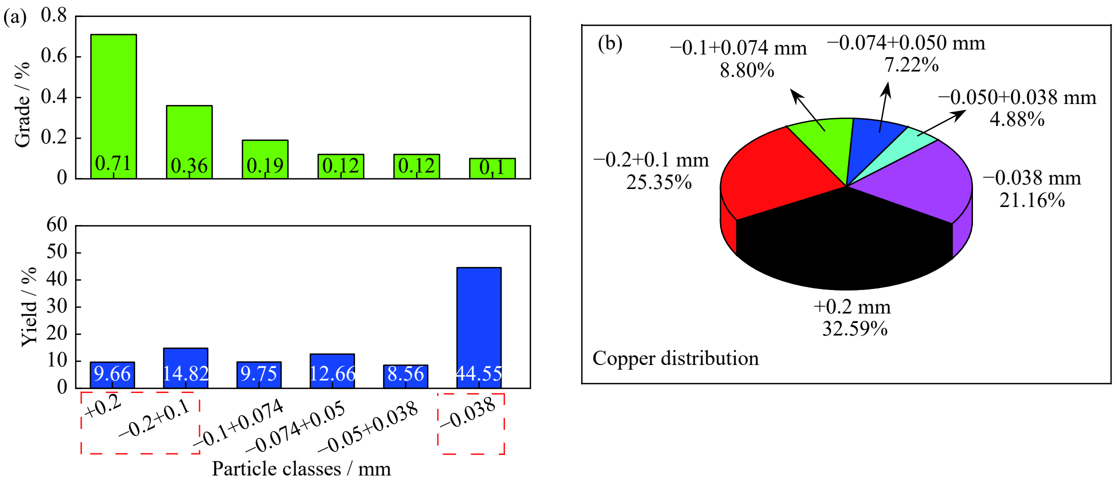

Fig. 2. Sieve analysis results of the flotation tailings: (a) grade and yield across different particle size fractions; (b) distribution of copper in each particle size fraction.

Table 2. Effect of regrinding process on the flotation performance.

Sample	Particle sizes	Products	Yield / %	Cu grade / %	Cu recovery / %
Unground tailings	+0.1 mm, 100wt%	Concentrate	3.61	3.80	30.78
		Tailings	96.39	0.32	69.22
		Feed ore	100.00	0.45	100.00
Reground tailings	−0.074 mm, 62wt%	Concentrate	3.59	10.25	74.59
		Tailings	96.41	0.13	25.41
		Feed ore	100.00	0.49	100.00

3.2. Mineralogical composition

The mineral composition and elemental deportment of the flotation feed and tailings samples from the production site were analyzed using XRD and MLA. The results indicate that the tailings sample contained significant amounts of quartz, orthoclase, and micas, as shown in Fig. 3. Characteristic peaks of chalcopyrite ($CuFeS_2$) were observed in the XRD pattern of the flotation feed, whereas these peaks were absent in the XRD pattern of the tailings sample. This is because the content of chalcopyrite is lower than the XRD detection limit of 0.2wt% in the copper tailings. Sracek *et al.* [48] found that chalcopyrite and bornite were predominant minerals disseminated in the Chambishi ore shale, but their contents were so low in the tailings that they could not be detected by XRD but only by more sophisticated detection methods. Considering this, MLA was employed to identify the mineral phase constituents of copper flotation tailings in this work. The results confirm that the tailings sample from the production site contained both primary and secondary copper sulfide minerals, and chalcopyrite was the dominant copper-bearing mineral phase. Jacobs [49] reported that chalcopyrite was the major mineral that occurred as fine-grained disseminations and banded structures at the Kansanshi deposit of Zambia, which provides solid support for our conclusion. Fig. 3(d) demonstrates that only slight variations existed in the abundances of gangue minerals before and after flotation, whereas the amounts of sulfide minerals significantly decreased after flotation, especially chalcopyrite and bornite. This phenomenon implies that most of the sulfide minerals in the flotation feed were recovered to concentrate products by froth flotation.

The deportments of Cu, Fe, and S elements across different minerals in the tailings are demonstrated in Fig. 4. The result shows that 72.83wt% Cu and 77.40wt% S were contributed by chalcopyrite, which further confirms that chalcopyrite was the dominant copper-containing mineral. Brochantite and Fe-bearing dioptase were the main non-sulfide phase minerals containing copper, but their weight percentages were less than 0.02wt%. Approximately 60wt% of the total iron occurred in the form of biotite, whereas the contribution of chalcopyrite only accounted for 19.85%. This is why the reduction of Fe concentration is not in accordance with those of Cu and S elements. Almost 75wt% of the total iron widely occurred in the non-sulfide phase minerals, such as biotite, hematite, and clinochlore.

3.3. Mineral grain size distribution

The flotation performance is strongly dependent on the degree of mineral liberation, which is directly related to the mineral grain size [50]. Several studies have reported that the flotation efficiency is relatively high under narrow range of particle size, approximately 20–150 μm [51–52]. Bahrami

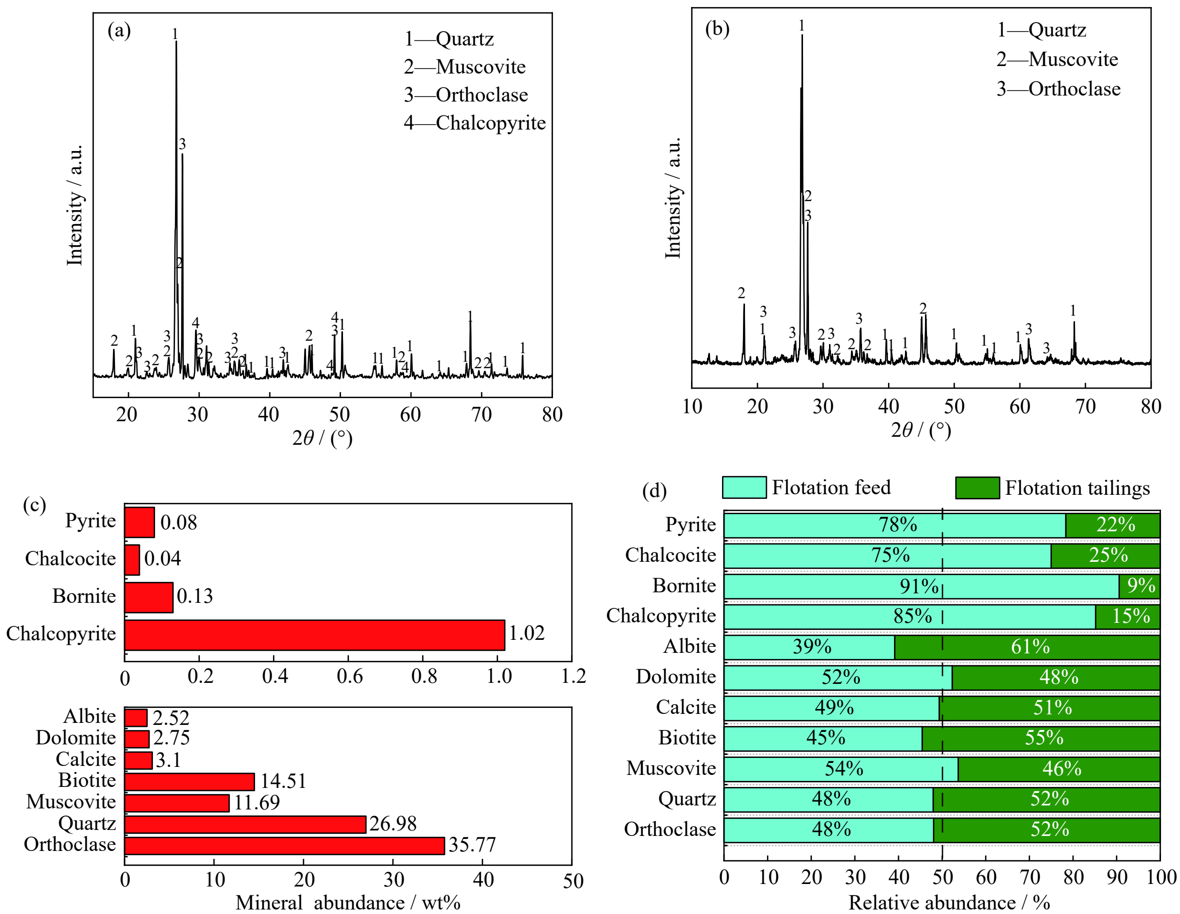

Fig. 3. Mineral constituents of the flotation feed and tailings samples: XRD patterns of the (a) flotation feed and (b) tailings sample; (c) modal mineralogy of the flotation tailings; (d) relative mineral abundance determined by MLA.

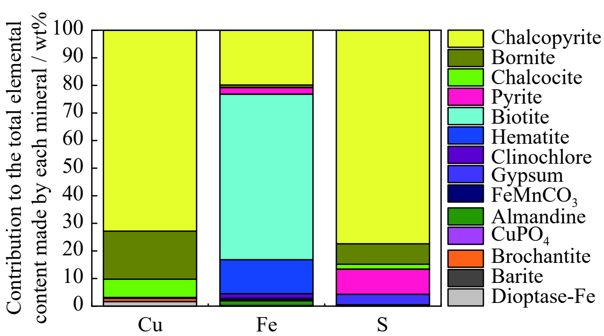

Fig. 4. Elemental deportment of Cu, Fe, and S across the main minerals in the flotation tailings.

et al. [53] found that the grade and recovery of concentrates increased with increasing particle size of chalcopyrite, and the maximum flotation rate occurred under the size range of 50–55 μm. In the current study, MLA measurement provides a quantitative description of the grain size distribution across different minerals. More detailed information on the grain size distribution across different minerals is presented in Fig. 5.

Interestingly, a significant difference in grain size distribution existed between the sulfide and gangue minerals. The grains of sulfide minerals were finer than those of gangue minerals, with the maximum grain size less than 180 μm. Almost all copper minerals were accumulated in the size fraction less than 106 μm, except for 9.2wt% chalcopyrite that occurred in the 160–180 μm size fraction. The amounts of gangue minerals retained in the size fractions larger than 106 μm were significantly higher than the amounts retained in the lower size fractions, and the proportions of orthoclase and muscovite even reached 60.31wt% and 51.76wt%, respectively. Compared with chalcopyrite and bornite, chalcocite exhibited the finest particle size distribution, and the particle size corresponding to the cumulative passing minerals of 80wt% (P_{80}) is 81.42 μm. There was uniform grain size distribution between chalcopyrite and bornite, indicating that most of them were locked together in the copper tailings. The MLA-derived cross-sectional image of the flotation tailings is shown in Fig. 5(d), in which mineral particles are sorted by particle area. The result further illustrates that copper minerals occurred in a highly dispersed state, and quite a few were fully encapsulated in coarse gangue particles. Hence, further grinding is required to liberate the locked copper minerals,

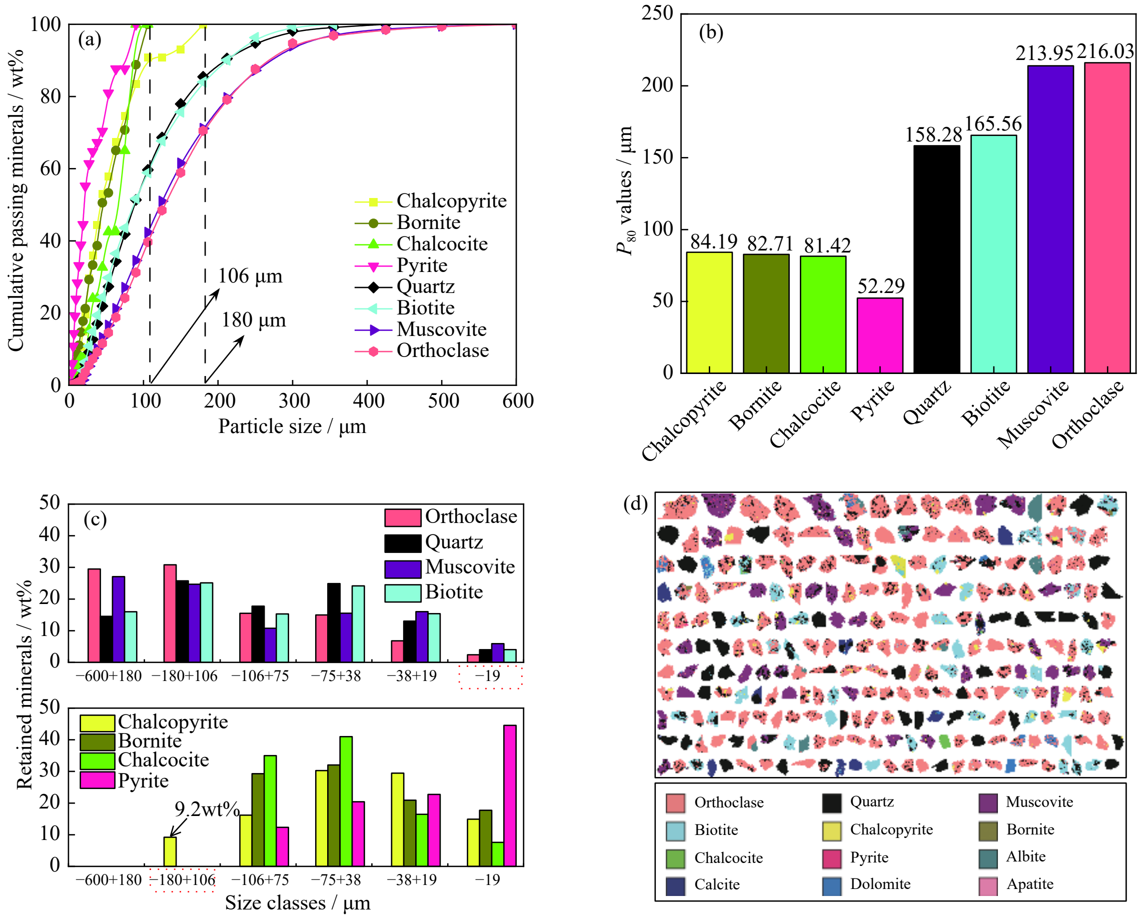

Fig. 5. Grain size distribution of different minerals in the flotation tailings: (a) cumulative weight percentage passing curve; (b) the particle size corresponding to the cumulative passing minerals of 80wt% (P_{80}); (c) weight percentage retained in each size class; (d) colored MLA-derived image sorted by particle area.

which will result in more energy consumption.

3.4. Mineral association and locking

To further understand the occurrence of copper minerals in the flotation tailings, SEM–EDS analysis was performed on the powder sample of tailings and the polished sections. Through contrastive analysis, copper minerals were found to occur in a highly dispersed state, and a few copper minerals grains were fully liberated from gangue minerals (Fig. 6). From the local SEM micrograph of a powder sample, a significant quantity of gangue minerals featured relatively coarse particle size, even larger than 150 μm. The EDS-mapping image of the powder sample demonstrated that it contained few copper minerals, with very fine granularity (less than 10 μm). In addition, some liberated pyrite grains were also present in the tailings. A few copper minerals were uncovered when the powder sample was polished, indicating that they were completely enclosed by gangue mineral particles, so that they could not be effectively recovered in the flotation process.

Fig. 7 demonstrates the association and locking profiles for sulfide minerals, including chalcopyrite, bornite, chalcocite, 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 (Fig. 7(a)). The free surfaces of chalcopyrite and bornite accounted for 20.76% and 19.72% of the total phase boundary, whereas the proportion of liberated chalcocite only reached 11.44%. This implies the liberation degree of chalcocite was relatively poorer than those of chalcopyrite and bornite. Additionally, copper minerals were closely associated with orthoclase and micas, especially orthoclase. About 54.94% of the chalcopyrite phase boundary length was associated with orthoclase phase boundary, 9.40% contacted with muscovite, 6.83% contacted with biotite, and only 3.61% was related to quartz.

The distribution of chalcopyrite, which has been identified as the main copper-bearing mineral across different liberation classes, is presented in Fig. 7(b). Orthoclase and muscovite were the predominant gangue minerals and were highly intergrown with unliberated chalcopyrite. The abundance of quartz in the unliberated chalcopyrite was signific-

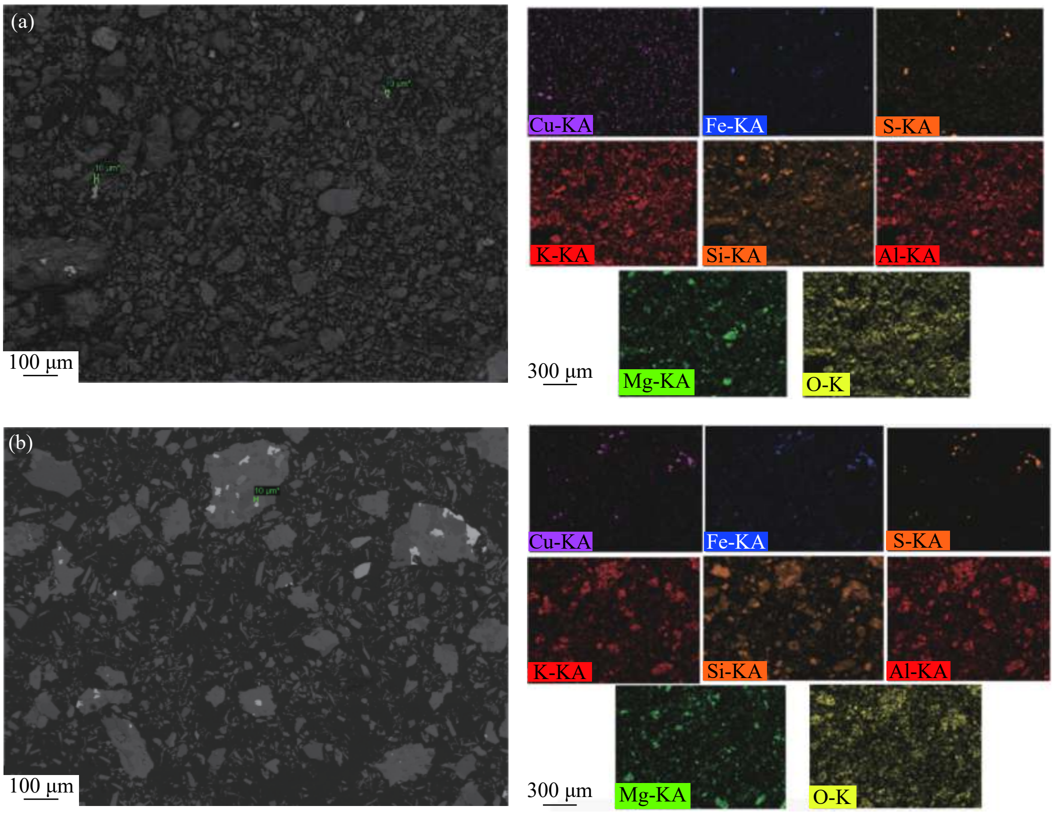

Fig. 6. SEM images and elemental mapping of copper flotation tailings: (a) powder sample; (b) polished sample.

antly lower than that in the copper-free liberated fraction. Quartz (SiO_2), belonging to the common rock-forming minerals, is considered the dominant gangue mineral in copper sulfide tailings [54]. Nevertheless, compared with orthoclase and micas, quartz was relatively low in the flotation tailings from the Chambishi Copper Mine. The phenomenon that feldspar occurs as the most abundant silicate minerals with a very little amount of quartz is always found in tailings from alkalic Cu-porphyry mine such as Mount Polley [55–56]. Therefore, the unusual mineral compositions are attributed to the genesis of copper mineralization in Chambishi areas and the geochemistry of copper tailings.

Furthermore, the displacement of mineral contents derived from the results of modal mineralogy and mineral association can be used as a powerful parameter to evaluate the breakage characteristics, as reported by Sandmann and Gutzmer [45]. According to the results of mineral association for chalcopyrite, bornite, and chalcocite, the orthoclase-to-quartz ratios were 15.22, 4.67, and 23.12, respectively. They are much higher than the expected value of 1.33 obtained from the modal mineralogy data. Therefore, copper minerals, particularly chalcocite, were highly intergrown with orthoclase rather than quartz, which is consistent with the abovementioned conclusion. For chalcopyrite, the orthoclase-to-muscovite ratio was 5.84 in the mineral association section, which is slightly larger than the expected value of 3.06 from the result of modal mineralogy. On the contrary, for bornite and chalcocite, the values were smaller: 1.60 and 2.82, respectively. This finding indicates that the chalcopyrite–orthoclase boundary breakage was relatively weaker than the chalcopyrite–muscovite boundary breakage, whereas bornite and chalcocite tended to be more associated with the muscovite phase boundary. In terms of mineral content, chalcopyrite was the predominant copper mineral, while the contents of bornite and chalcocite only accounted for 0.13wt% and 0.04wt% (Fig. 3(c)). As shown in Figs. 7(c) and 7(d), 81.57wt% chalcopyrite and 84.10wt% bornite were locked up in multiphase particles, and the uniform mineral distribution indicates that they were often locked with each other in the tailings. The fully liberated chalcopyrite and bornite only accounted for 3.28wt% and 3.13wt% (Fig. 7(c)), whereas liberated chalcocite grains could not be found in the flotation tailings. Furthermore, orthoclase was always the most abundant gangue mineral no matter the type of locking relationship. Compared with chalcopyrite and bornite, the occurrence mode of chalcocite was a bit more complex. Overall, the copper minerals were highly dispersed in the form of fine grains, and some were even completely locked up in multiphase gangue particles.

3.5. Mineral liberation measurements

Fig. 8 presents the liberation characteristics of major copper minerals in the form of histograms and curves, which were computed based on the exposed free surface. As expec-

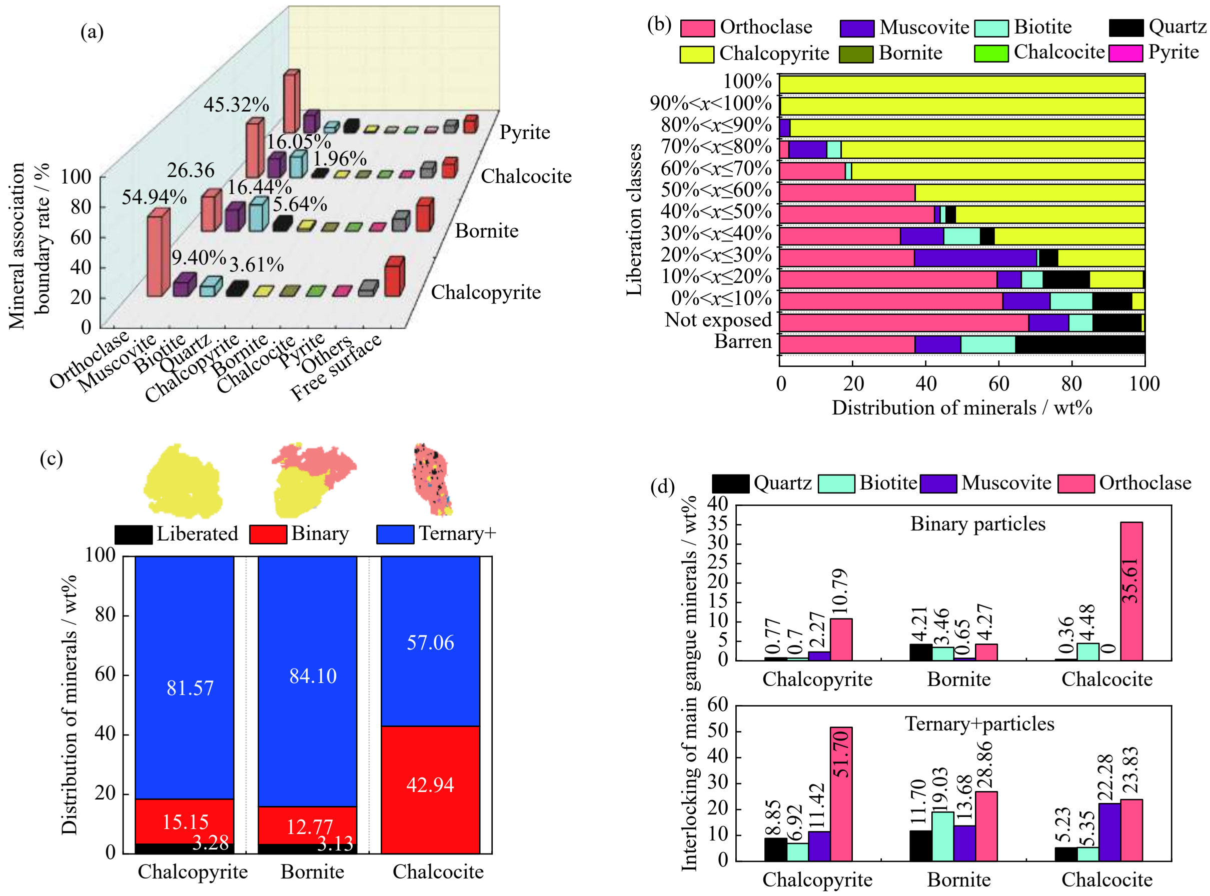

Fig. 7. Mineral association and locking profile of the flotation tailings: (a) sulfide minerals association with gangue minerals; (b) chalcopyrite liberation characteristics; (c, d) copper sulfide minerals locking and the contiguous relationship between major gangue minerals.

ted, the degree of liberation of copper minerals markedly became worse after the flotation process. Moreover, 85.10% chalcopyrite and 82.6% bornite were accumulated in the <30% liberation classes, which suggests a lean intergrowth of copper-hosting minerals and gangue fractions, and the distribution of chalcocite in this fraction even reached 100%. For chalcopyrite and bornite, 8.88% and 13.96% were fully locked within copper-free minerals, respectively, whereas for chalcocite, the value rises to 32.25%. It is inferred from the results that copper minerals exhibited an extremely poor degree of liberation in the tailings, and among them, chalcocite had the worst liberation degree. The theoretical cumulative recovery–grade curve in Fig. 8(c) reflects the maximum expected recovery by flotation at a fixed grade, which is essentially dependent on the mineralogy and texture [57–59]. The overall curve is pushed to the top-right corner, which indicates that the mineralogy properties of the sample have been changed and the liberation of valuable minerals has become better. Compared with the flotation feed, the chalcopyrite and bornite in the tailings sample resulted in a lower grade–recovery curve (Fig. 8(c)); this is because they were finely disseminated in the gangue mineral particles and remained as locked structures in the flotation tailings.

The selected MLA image of the tailings sample from the production site, in which particles are sorted by chalcopyrite free boundary, is presented in Fig. 9. The cross-sectional image further illustrates that chalcopyrite was extremely finely dispersed and wrapped in gangue mineral particles. Although the sample contained a few liberated chalcopyrite particles, the area percentage only accounted for 3.25% of the total mineral area. The particle size of the liberated chalcopyrite was less than 45 μm, whereby more than 80% had a particle size less than 20 μm. For sulfide minerals, the optimal granularity range is between 30 and 150 μm in the industry, as reported by Mankosa *et al.* [60]. Several previous studies have reported that most of the copper losses in the flotation tailings occur in particle size fractions less than 20 μm (50%) and larger than 150 μm (30%–40%) [61–62]. Asghari *et al.* [15] found that the most sulfide copper losses occurred in coarse particles (>74 μm) and that the most oxide copper loss occurred in fine particles (<9 μm). As a result, liberated chalcopyrite with particle size less than 20 μm has a high probability to be lost to the tailings stream because of its ultrafine granularity and inherent limitations related to phys-

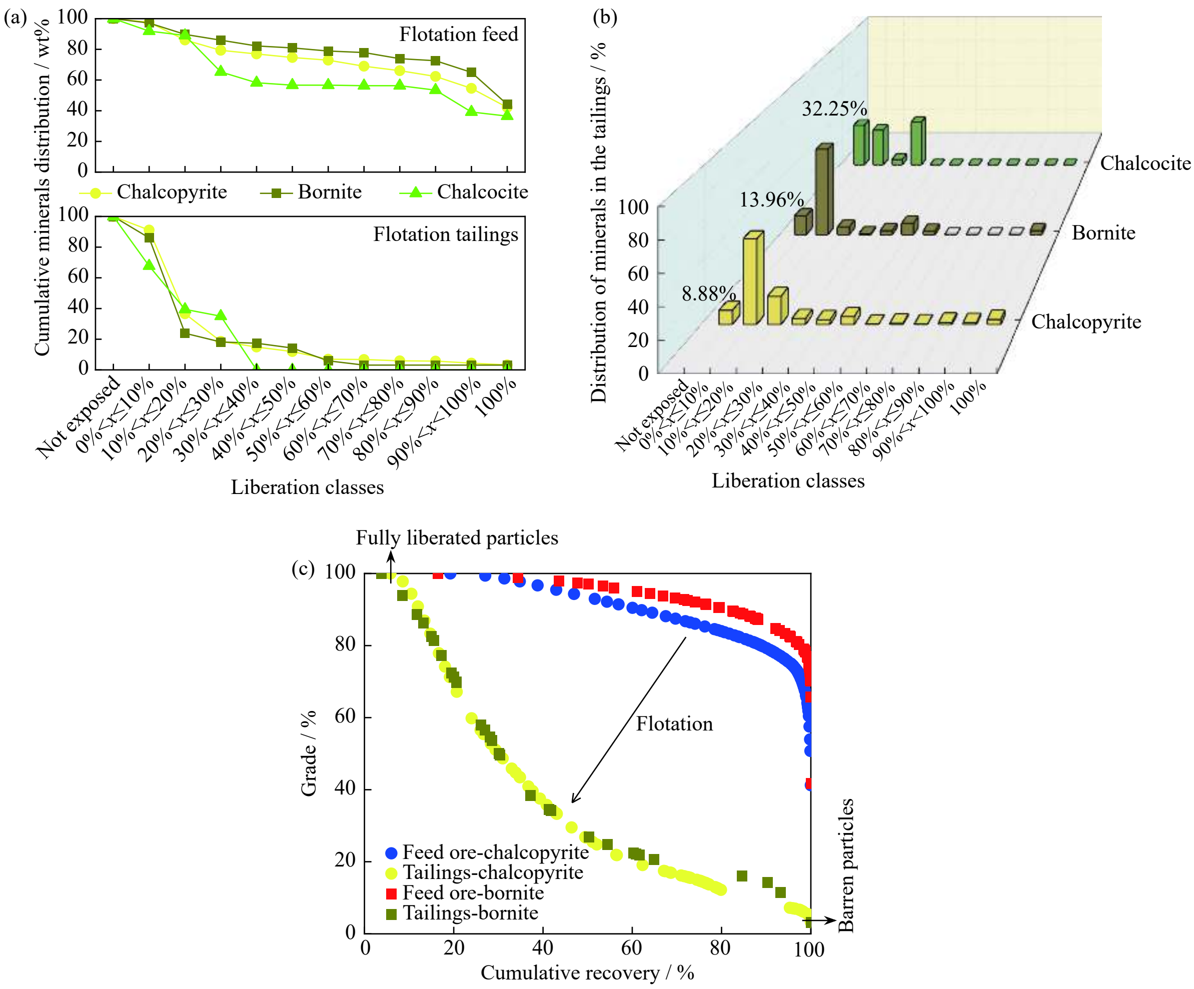

Fig. 8. Copper mineral liberation based on the free surface of particles before and after flotation: (a) cumulative distribution in various degrees; (b) distribution of minerals retained across different liberation classes; (c) theoretical cumulative recovery versus grade.

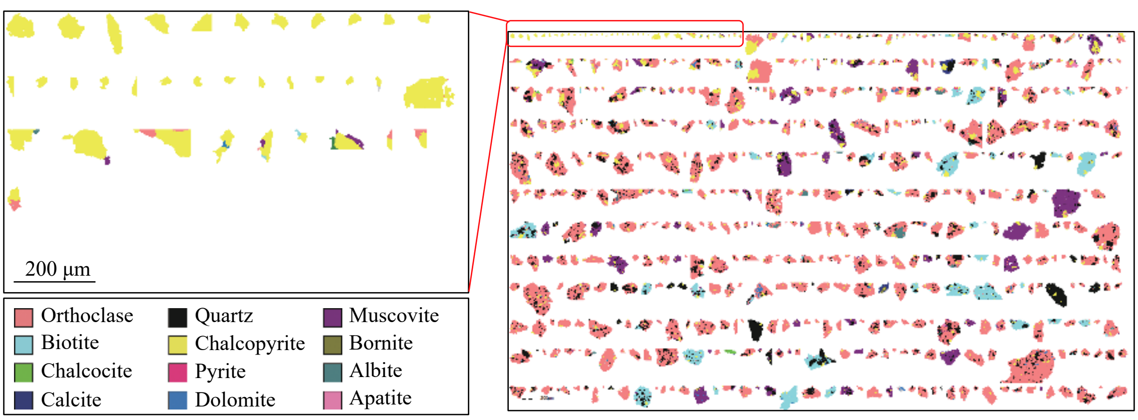

Fig. 9. Selected MLA image of copper tailings sorted by chalcopyrite free boundary.

ical interactions.

4. Conclusions

In this study, a comprehensive mineralogical characterization was carried out using quantitative MLA integrated with SEM–EDS to further clarify the occurrence mode of copper in flotation tailings from the Chambishi Copper Mine. The SEM–EDS comparative analysis of the powder sample and polished sample confirmed that most of the copper minerals were completely enclosed by gangue minerals. The unrecovered copper in the tailings occurred in a few liberated ultrafine particles and interlocked structures; these structures which were mainly highly dispersed and completely enclosed. The conclusions drawn from this research are as follows:

(1) Chalcopyrite was the dominant copper mineral, accounting for approximately 72.83% of the total copper, and it was highly intergrown with orthoclase and muscovite rather than quartz.

(2) The phase boundary breakage of chalcopyrite–orthoclase was relatively weaker than that of chalcopyrite–muscovite, whereas bornite and chalcocite tended to be more associated with the muscovite phase boundary

(3) The liberated chalcopyrite grains, only accounting for 3.28wt%, remained unrecovered in the flotation tailings because of their ultrafine particle size. Approximately 85% of chalcopyrite was distributed in the <30% liberation classes, indicating that the liberation of unrecovered copper minerals requires a further reduction in particle size.

(4) The grain size distribution of sulfide minerals was significantly finer than that of gangue minerals, with the maximum grain size of the former less than 180 μm. Almost all copper minerals were distributed in the size fractions less than 106 μm, except for 9.2wt% chalcopyrite in the 160–180 μm size fraction.

(5) Copper minerals predominantly occurred as fine-grained disseminations within gangue minerals. The area percentages of chalcopyrite and bornite completely encapsulated by gangue minerals were 8.88% and 13.96%, respectively, whereas the proportion reached 32.25% for locked chalcocite.

Acknowledgements

This work was financially supported by a grant from Nonferrous Corporation Africa Mining Public Limited Company and National Natural Science Foundation of China (No. 51804020). We gratefully acknowledge NFC Africa Mining Plc. for sample collection and delivery. We also would like to acknowledge Institute of Process Engineering, Chinese Academy of Sciences for the MLA measurement.

References

[1] M.E. Schlesinger, M.J. King, K.C. Sole, and W.G. Davenport, *Extractive Metallurgy of Copper*, Elsevier, Amsterdam, 2011.

[2] K.J. Nyembwe, E. Fosso-Kankeu, F. Waanders, and K.D. Nyembwe, Structural, compositional and mineralogical characterization of carbonatitic copper sulfide: Run of mine, concentrate and tailings, *Int. J. Miner. Metall. Mater.*, 26(2019), No. 2, p. 143.

[3] Y. Li, N. Kawashima, J. Li, A.P. Chandra, and A.R. Gerson, A review of the structure. and fundamental mechanisms and kinetics of the leaching of chalcopyrite, *Adv. Colloid Interface Sci.*, 197-198(2013), p. 1.

[4] D. Mesa and P.R. Brito-Parada, Scale-up in froth flotation: A state-of-the-art review, *Sep. Purif. Technol.*, 210(2019), p. 950.

[5] B. Feng, C.H. Zhong, L.Z. Zhang, Y.T. Guo, T. Wang, and Z.Q. Huang, Effect of surface oxidation on the depression of sphalerite by locust bean gum, *Miner. Eng.*, 146(2020), art. No. 106142.

[6] M.M. Antonijević, M.D. Dimitrijević, Z.O. Stevanović, S.M. Serbula, and G.D. Bogdanovic, Investigation of the possibility of copper recovery from the flotation tailings by acid leaching, *J. Hazard. Mater.*, 158(2008), No. 1, p. 23.

[7] B.S. Han, B. Altansukh, K. Haga, Z. Stevanović, R. Jonović, L. Avramović, D. Urosević, Y. Takasaki, N. Masuda, D. Ishiyama, and A. Shibayama, Development of copper recovery process from flotation tailings by a combined method of high-pressure leaching-solvent extraction, *J. Hazard. Mater.*, 352(2018), p. 192.

[8] J. Alcalde, U. Kelm, and D. Vergara, Historical assessment of metal recovery potential from old mine tailings: A study case for porphyry copper tailings, Chile, *Miner. Eng.*, 127(2018), p. 334.

[9] Z.G. Yin, W. Sun, Y.H. Hu, C.H. Zhang, Q.J. Guan, and K.P. Wu, Evaluation of the possibility of copper recovery from tailings by flotation through bench-scale, commissioning, and industrial tests, *J. Cleaner Prod.*, 171(2018), p. 1039.

[10] J. Pazhoohan, H. Beiki, and M. Esfandyari, Experimental investigation and adaptive neural fuzzy inference system prediction of copper recovery from flotation tailings by acid leaching in a batch agitated tank, *Int. J. Miner. Metall. Mater.*, 26(2019), No. 5, p. 538.

[11] S.H. Yin, L.M. Wang, A.X. Wu, E. Kabwe, X. Chen, and R.F. Yan, Copper recycle from sulfide tailings using combined leaching of ammonia solution and alkaline bacteria, *J. Cleaner Prod.*, 189(2018), p. 746.

[12] K. Nakajima, I. Daigo, K. Nansai, K. Matsubae, W. Takayanagi, M. Tomita, and Y. Matsuno, Global distribution of material consumption: Nickel, copper, and iron, *Resour. Conserv. Recycl.*, 133(2018), p. 369.

[13] H.K. Hansen, J.B. Yianatos, L.M. Ottosen, Speciation and leachability of copper in mine tailings from porphyry copper mining: influence of particle size, *Chemosphere*, 60(2005), No. 10, p. 1497.

[14] S. Jannesar Malakooti, S.Z. Shafaei Tonkaboni, M. Noaparast, F. Doulati Ardejani, and R. Naseh, Characterisation of the Sarcheshmeh copper mine tailings, Kerman province, southeast of Iran, *Environ. Earth Sci.*, 71(2014), No. 5, p. 2267.

[15] M. Asghari, F. Nakhaei, and O. VandGhorbany, Copper recovery improvement in an industrial flotation circuit: A case study of Sarcheshmeh copper mine, *Energy Sources Part A*, 41(2019), No. 6, p. 761.

[16] I. Mackay, E. Mendez, I. Molina, A.R. Videla, J.J. Cilliers, and P.R. Brito-Parada, Dynamic froth stability of copper flotation tailings, *Miner. Eng.*, 124(2018), p. 103.

[17] R.F. Zhu, G.H. Gu, Z.X. Chen, Y.H. Wang, and S.Y. Song, A new collector for effectively increasing recovery in copper oxide ore-staged flotation, *Minerals*, 9(2019), No. 10, p. 595.

[18] B. Feng, W.P. Zhang, Y.T. Guo, J.X. Peng, X.H. Ning, and H.H. Wang, Synergistic effect of acidified water glass and locust bean gum in the flotation of a refractory copper sulfide ore, *J. Cleaner Prod.*, 202(2018), p. 1077.

[19] S. Northey, S. Mohr, G.M. Mudd, Z. Weng, and D. Giurco, Modelling future copper ore grade decline based on a detailed

assessment of copper resources and mining, *Resour. Conserv. Recycl.*, 83(2014), p. 190.

[20] B. Golding and S.D. Golding, *Metals, Energy and Sustainability*, Springer, Cham, 2017, p. 21.

[21] R.B. Gordon, Production residues in copper technological cycles, *Resour. Conserv. Recycl.*, 36(2002), No. 2, p. 87.

[22] C.C. Lü, Y.L. Wang, P. Qian, Y. Liu, G.Y. Fu, J. Ding, S.F. Ye, and Y.F. Chen, Separation of chalcopyrite and pyrite from a copper tailing by ammonium humate, *Chin. J. Chem. Eng.*, 26(2018), No. 9, p. 1814.

[23] A.C. Brown, World-class sediment-hosted stratiform copper deposits: Characteristics, genetic concepts and metallotects, *Aust. J. Earth Sci.*, 44(1997), No. 3, p. 317.

[24] R.R. McGowan, S. Roberts, and A.J. Boyce, Origin of the Nchanga copper—Cobalt deposits of the Zambian Copperbelt, *Miner. Deposita*, 40(2006), No. 6, p. 617.

[25] J.L.H. Cailteux, A.B. Kampunzu, C. Lerouge, A.K. Kaputo, and J.P. Milesi, Genesis of sediment-hosted stratiform copper-cobalt deposits, central African Copperbelt, *J. Afr. Earth Sci.*, 42(2005), No. 1-5, p. 134.

[26] J.K. Wen, B.W. Chen, H. Shang, and G.C. Zhang, Research progress in biohydrometallurgy of rare metals and heavy nonferrous metals with an emphasis on China, *Rare Met.*, 35(2016), No. 6, p. 433.

[27] X.D. Hao, X.D. Liu, Q. Yang, H.W. Liu, H.Q. Yin, G.Z. Qiu, and Y.L. Liang, Comparative study on bioleaching of two different types of low-grade copper tailings by mixed moderate thermophiles, *Trans. Nonferrous Met. Soc. China*, 28(2018), No. 9, p. 1847.

[28] J.B. Chen, R.J. Li, and L.H. Yu, The way of the survey and assessment of copper tailings resources and their application, *J. Nat. Resour.*, 27(2012), No. 8, p. 1373.

[29] L. Pérez-Barnuevo, E. Pirard, and R. Castroviejo, Automated characterisation of intergrowth textures in mineral particles. A case study, *Miner. Eng.*, 52(2013), p. 136.

[30] Y. Gu, R.P. Schouwstra, and C. Rule, The value of automated mineralogy, *Miner. Eng.*, 58(2014), p. 100.

[31] R. Sousa, B. Simons, K. Bru, A.B. de Sousa, G. Rollinson, J. Andersen, M. Martin, and M.M. Leite, Use of mineral liberation quantitative data to assess separation efficiency in mineral processing—Some case studies, *Miner. Eng.*, 127(2018), p. 134.

[32] P.Y. Zhang, L.M. Ou, L.M. Zeng, W.G. Zhou, and H.T. Fu, MLA-based sphalerite flotation optimization: Two-stage roughing, *Powder Technol.*, 343(2019), p. 586.

[33] L. Liu, Q. Tan, L. Liu, and J.C. Cao, Comparison of different comminution flowsheets in terms of minerals liberation and separation properties, *Miner. Eng.*, 125(2018), p. 26.

[34] T. Leißner, K. Bachmann, J. Gutzmer, and U.A. Peuker, MLA-based partition curves for magnetic separation, *Miner. Eng.*, 94(2016), p. 94.

[35] Z. Li, Y.H. Fu, C. Yang, W. Yu, L.J. Liu, J.Z. Qu, and W. Zhao, Mineral liberation analysis on coal components separated using typical comminution methods, *Miner. Eng.*, 126(2018), p. 74.

[36] D.H. Hoang, A. Hassanzadeh, U.A. Peuker, and M. Rudolph, Impact of flotation hydrodynamics on the optimization of fine-grained carbonaceous sedimentary apatite ore beneficiation, *Powder Technol.*, 345(2019), p. 223.

[37] C.L. Xu, C.B. Zhong, R.L. Lyu, Y.Y. Ruan, Z.Y. Zhang, and R.A. Chi, Process mineralogy of Weishan rare earth ore by MLA, *J. Rare Earths*, 37(2019), No. 3, p. 334.

[38] B. Babel, M. Penz, E. Schach, S. Boehme, and M. Rudolph, Reprocessing of a southern Chilean Zn tailing by flotation—A case study, *Minerals*, 8(2018), No. 7, p. 295.

[39] K. Berkh, D. Rammlmair, M. Drobe, and J. Meima, Case study: Geochemistry and mineralogy of copper mine tailings in Northern Central-Chile, [in] *14th International Congress for Applied Mineralogy*, Belgorod, 2019, p. 37..

[40] B. Forsyth, M. Edraki, and T. Baumgartl, The evolution of tailings seepage chemistry at one of Australia's largest and longest operating mines, [in] *10th International Conference on AcidRock Drainage and Annual IMWA Conference*, Santiago, 2015, p. 1.

[41] L. Wang and M.Y. Wang, Research on copper dissemination state from old tailings in a copper mine, *Nonferrous Met. (Miner. Process. Sect.)*, 2012, No. 6, p. 1.

[42] D. Naumov, L. Stamenov, S. Gaydardzhiev, and H. Bouzahzah, Coupling mineralogy with physicochemical parameters in view copper flotation efficiency improvement, *Physicochem. Probl. Miner. Process.*, 55(2019), No. 3, p. 701.

[43] M. Lu, D.H. Xie, W.H. Gui, L.H. Wu, C.Y. Chen, and C.H. Yang, A cascaded recognition method for copper rougher flotation working conditions, *Chem. Eng. Sci.*, 175(2018), p. 220.

[44] F. Kukurugya, A. Rahfeld, R. Möckel, P. Nielsen, L. Horckmans, J. Spooren, and K. Broos, Recovery of iron and lead from a secondary lead smelter matte by magnetic separation, *Miner. Eng.*, 122(2018), p. 17.

[45] D. Sandmann and J. Gutzmer, Use of mineral liberation analysis (MLA) in the characterization of lithium-bearing micas, *J. Miner. Mater. Charact. Eng.*, 1(2013), No. 6, p. 285.

[46] B. Schulz, G. Merker, and J. Gutzmer, Automated SEM mineral liberation analysis (MLA) with generically labelled EDX spectra in the mineral processing of rare earth element ores, *Minerals*, 9(2019), No. 9, p. 527.

[47] A. Bakalarz, M. Duchnowska, and A. Luszczkiewicz, Influence of liberation of sulphide minerals on flotation of sedimentary copper ore, [in] *E3S Web of Conferences*, Paris, 2017, art. No. 01025.

[48] O. Sracek, M. Mihaljevič, B. Kříbek, V. Majer, and F. Veselovský, Geochemistry and mineralogy of Cu and Co in mine tailings at the Copperbelt, Zambia, *J. Afr. Earth Sci.*, 57(2010), No. 1-2, p. 14.

[49] T.T. Jacobs, *Process Mineralogical Characterisation of the Kansanshi Copper Ore, NW Zambia* [Dissertation], University of Cape Town, Cape Town, 2016.

[50] P. Vallejos, J. Yianatos, L. Vinnett, and L. Bergh, Characterization of the industrial flotation process based on size-liberation relationships, *Miner. Eng.*, 121(2018), p. 189.

[51] A. Norori-McCormac, P.R. Brito-Parada, K. Hadler, K. Cole, and J.J. Cilliers, The effect of particle size distribution on froth stability in flotation, *Sep. Purif. Technol.*, 184(2017), p. 240.

[52] A.M. Gaudin, R. Schuhmann Jr., and A.W. Schlechten, Flotation kinetics. II. The effect of size on the behavior of galena particles, *J. Phys. Chem.*, 46(1942), No. 8, p. 902.

[53] A. Bahrami, M. Mirmohammadi, Y. Ghorbani, F. Kazemi, M. Abdollahi, and A. Danesh, Process mineralogy as a key factor affecting the flotation kinetics of copper sulfide minerals, *Int. J. Miner. Metall. Mater.*, 26(2019), No. 4, p. 430.

[54] D. Kossoff, W.E. Dubbin, M. Alfredsson, S.J. Edwards, M.G. Macklin, and K.A. Hudson-Edwards, Mine tailings dams: Characteristics, failure, environmental impacts, and remediation, *Appl. Geochem.*, 51(2014), p. 229.

[55] S. Hashmi, B. Ward, A. Plouffe, T. Ferbey, and M. Leybourne, Geochemical and mineralogical dispersal in till from the Mount Polley Cu–Au porphyry deposit, central British Columbia,

Canada, *Geol. Surv. Can.*, (2014), art. No. 7589.

[56] C. Kennedy, S.J. Day, and C.D. Anglin, Geochemistry of tailings from the Mount Polley Mine, British Columbia, [in] *Proceedings Tailings and Mine Waste 2016*, Keystone, 2016, p. 857.

[57] A.F. Cropp, W.R. Goodall, and D.J. Bradshaw, The influence of textural variation and gangue mineralogy on recovery of copper by flotation from porphyry ore—A review, [in] *The Second AusIMM International Geometallurgy Conference*, Brisbane, 2013, p.279.

[58] C.B. Zhong, C.L. Xu, R.L. Lyu, Z.Y. Zhang, X.Y. Wu, and R.A. Chi, Enhancing mineral liberation of a Canadian rare earth ore with microwave pretreatment, *J. Rare Earths*, 36(2018), No. 2, p. 215.

[59] M. Camalan, M. Çavur, and Ç. Hoşten, Assessment of chromite liberation spectrum on microscopic images by means of a supervised image classification, *Powder Technol.*, 322(2017), p. 214.

[60] M.J. Mankosa, J.N. Kohmuench, L. Christodoulou, and G.H. Luttrell, Recovery of values from a porphory copper tailings stream, [in] *The XXVIII International Mineral Processing Congress*, Quebec, 2016, p. 11.

[61] S. Agheli, A. Hassanzadeh, B.V. Hassas, and M. Hasanzadeh, Effect of pyrite content of feed and configuration of locked particles on rougher flotation of copper in low and high pyritic ore types, *Int. J. Min. Sci. Technol.*, 28(2018), No. 2, p. 167.

[62] S.M. Bulatovic, D.M. Wyslouzil, and C. Kant, Operating practices in the beneficiation of major porphyry copper/molybdenum plants from Chile: Innovated technology and opportunities, a review, *Miner. Eng.*, 11(1998), No. 4, p. 313.