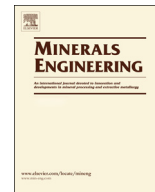




ELSEVIER

Contents lists available at ScienceDirect

Minerals Engineering

journal homepage: www.elsevier.com/locate/mineng

Historical assessment of metal recovery potential from old mine tailings: A study case for porphyry copper tailings, Chile

J. Alcalde^{a,b,*}, U. Kelm^a, D. Vergara^c

^a Instituto de Geología Económica Aplicada, Universidad de Concepción, Chile

^b Institut für Mineralogie, Technische Universität Bergakademie (TUBA) Freiberg, Germany

^c Departamento de Ingeniería de Minas, Universidad de Chile, Chile

ARTICLE INFO

Keywords:

Copper tailings reprocessing
Flotation
Selectivity and recovery
Theoretical grade-recovery curves
Copper speciation

ABSTRACT

The metal mining activity has been the source of large volumes of tailings that must be stored in tailings storage facilities (TSFs). Since tailings might contain residual valuable metals, these are often subject of assessment studies aimed to find a feasible way to reprocessing. One of these TSFs was built and operated during the 1930's for the exploitation of a giant porphyry copper deposit in Chile, a deposit that is still under exploitation today. For these old tailings a historical assessment was varied out about the copper recovery potential in a reprocessing scenario by flotation. Based on theoretical concepts and available historical operational data, it can be shown that the main reason for the relatively high copper grades found in these tailings are the corresponding high grades of the processed ores, instead of being the result of an inefficient past technology of flotation with consequential low recoveries. Conversely, the former flotation circuit was optimal concerning the recovery of copper sulphides favoured by the less restrictive high copper content of these sulphides. Consistently, it is argued for the study case that the main advances in flotation circuits have focused on improving selectivity without sacrificing recovery, due to the changes in the mineralogy of the exploited ores. These are relevant facts for a reprocessing scenario, indicating a low copper recovery potential if the current conventional flotation technology is considered; thus other and/or newer processing technologies could be more efficient for recovery of the remaining copper. There are hydrometallurgical potential alternatives for reprocessing but limitations are expected in relation to milling size of the tailings material. The present findings may apply elsewhere, due to the high representativeness of the present study case.

1. Introduction

The mining activity is and has been vital for the economy of many countries, e.g. Chile, especially since the beginning of the 20th century. It has resulted not only in the production of metals but also in the generation of waste products, particularly when processing sulphides ores from porphyry copper deposits. This leads to large amounts of tailings that must be stored in tailings storage facilities (TSFs) (Hansen et al., 2005). These mine tailings are the remaining fine grained (1–600 μm) ground-up rock after the minerals of economic importance have been extracted, mixed with the associated process water which includes dissolved metals and processing reagents (Edraki et al., 2014). Since the solid phase from tailings might contain remaining valuable metals that were not recovered in the past, they represent potential resources and therefore they are objects of assessment studies and eventually of reprocessing.

Reprocessing implies that the tailings material is used as a feedstock

for producing valuable products such as recovered minerals and metals (Edraki et al., 2014). According to Gordon (2002), mine tailings can be observed as the single largest source of copper in waste deposits in the US copper cycle and Johansson et al. (2013) categorized tailings as valuable stocks of metals in the “technosphere”, indicating that reprocessing could also be an innovative reclamation technology. The reprocessing of tailings for recovery of metals like gold (Bugnosen, 2001), copper (EPA, 1993) or iron (Karlberg, 2010) is a past and current practice. Rampacek (1982) provides an overview of mining and mineral processing waste as a resource; in 1994 for example, 2% of the total worldwide copper production came from reworked tailings (Graedel et al., 2004). In Chile since the early nineties mining companies have been established and exclusively dedicated to reprocess old as well as fresh porphyry copper tailings to recover copper and molybdenum.

Reprocessing of old tailings that contain significant amounts of copper could result in a higher rate of production than processing of

* Corresponding author at: Instituto de Geología Económica Aplicada, Universidad de Concepción, Chile.
E-mail address: juanalcalde.e@hotmail.com (J. Alcalde).

<https://doi.org/10.1016/j.mineng.2018.04.022>

Received 30 May 2017; Received in revised form 18 April 2018; Accepted 21 April 2018
0892-6875/ © 2018 Elsevier Ltd. All rights reserved.

primary ores (Edraki et al., 2014), especially considering that they were already mined and processed (Lutandula and Maloba, 2013). However, if the copper sulphides minerals are finely disseminated and intergrown with pyrite and associated or occluded with gangue minerals re-processing may not be feasible due to the high costs involved in both separation and liberation (Kitobo et al., 1999). In addition, the resulting speciation of the remaining copper from tailings and possible chemical changes that might affect the TSF “ore” body since its deposition will condition any recovery process (Dold and Fontboté, 2001; Edraki et al., 2014, Hansen et al., 2005).

Regarding the reason behind the relatively high copper grades that could be found in old tailings some authors have argued that this might be due to a past inefficient recovery technology (Dold and Weibel, 2013; Edraki et al., 2014; Falagán et al., 2017). For such scenario, re-processing could be a winning bet considering the current available technology, but the underlying reason might not be the lower past recoveries. In fact, according to Gordon (2002) the average worldwide copper recovery achieved by flotation technology has not changed since its beginning in the 1920’s. Therefore, a particular and deeper analysis should be performed since finding the reason behind the higher grades could be the starting point in any future reprocessing design.

It is a current practice to assess the metal recovery potential from tailings through the amount and grades of the stored material in combination with metallurgical testwork campaigns, a time - and money - consuming venture. The purpose of this work is performing such assessments for old porphyry copper tailings from Chile in a reprocessing scenario by flotation, only based on specially developed theoretical concepts and available historical operation data as an economically efficient first step of a reprocessing or even remediation initiative.

2. Assessment approach

The methodology combines theoretical concepts developed in this work with the available historical operation data. This combination allows a focused interpretation of the data and thus, an assessment of the copper recovery potential from the studied old tailings.

2.1. Theoretical concepts

Three concepts are highlighted in the following sections as a theoretical base for the assessment of the available historical data.

2.1.1. Commitment between selectivity and recovery

This concept is related to the inherent commitment between the concentrate grade, as a measure of selectivity, and the recovery that both take place in the concentration process by flotation. This commitment becomes clearer when a theoretical grade-recovery curve is used, because it is a way to visualize the trade-off between the two main objectives of flotation, i.e., achieving a high concentrate grade while sacrificing as little recovery as possible (Neethling and Cilliers, 2012). This curve is referred to the combinations of maximal values of concentrate grades and recoveries that a flotation circuit can ever reach limited by feed ore mineralogy and textural aspects after grinding, i.e., liberation degree. Fig. 1 shows a comparison between two hypothetical cases using typical theoretical grade-recovery curves considering in both cases the same textural aspects after grinding.

Chalcocite with a copper content of 79.9% and chalcopyrite with a lower content of 34.6% are considered for this comparison, as they reflect the change in ore mineralogy of typical porphyry deposits throughout the aging of the exploitation, especially of those with secondary enrichment zones.

2.1.2. Mass balance equations (origin of tailings copper grade)

This second concept is related to the origin of the copper grades with which tailings end up after flotation. Even though it is obvious that the corresponding ore deposit and the specific mine sites and processing

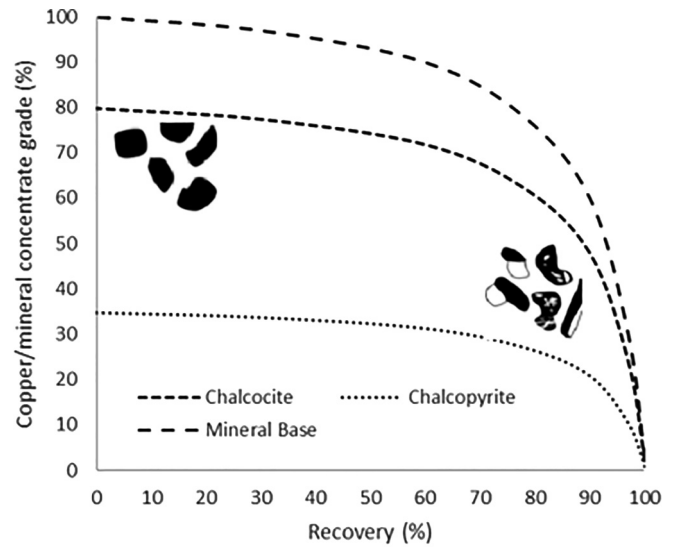


Fig. 1. Typical theoretical grade-recovery curves on a copper and mineral base.

conditions determine this copper content, a focus is needed on the following mass and copper balance equations that govern the concentration process by flotation.

$$F = C + T \quad (1)$$

$$FG_f = CG_c + TG_t \quad (2)$$

The variables F, C and T are the total mass of the feed ore, the concentrate and the tailings, and Gf, Gc and Gt are the corresponding copper grades of the same fluxes.

These two simple mass balance equations are the basis of any metallurgical analysis of a concentration process like flotation. For instance, the recovery (Rec) can be expressed in terms of the feed ore grade and the concentrate and tailings grades reached by the process, as the former two equations lead to the following equation.

$$Rec = \frac{G_c(G_f - G_t)}{G_f(G_c - G_t)} \quad (3)$$

Based on the Eq. (3) an expression for the copper grade in tailings also can be written as follows.

$$G_t = \frac{G_c \cdot G_f \cdot (1 - Rec)}{G_c - G_f \cdot Rec} \quad (4)$$

Eq. (4) represents an expression for the calculation of the tailings grade based on the feed ore grade and the two main objectives of the process: recovery and concentrate grade. Nevertheless, a more detailed analysis of Eq. (4) shows that for the tailings grade there are practically no changes versus changes in the concentrate grade above certain common limits ($\frac{\partial G_t}{\partial G_c} \approx 0$; for $G_c > 15\%$) as is shown in Fig. 2 for five

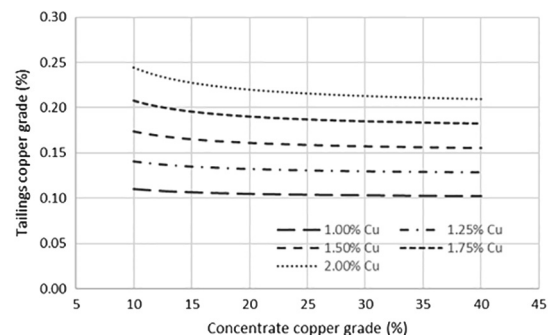


Fig. 2. Changes on tailings grades with the concentrate grades.

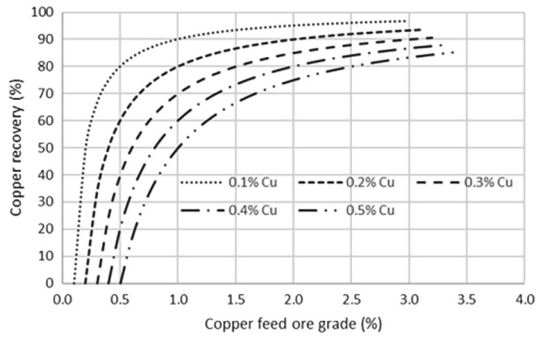


Fig. 3. Iso-grades curves of copper in tailings.

given feed ore grades and considering a 90% of copper recovery.

Moreover, the expression $\left(\frac{G_c}{G_c - G_{f-Rec}}\right)$ is the inverse of the mass pull related to tailings and is commonly close to 1, reflecting the nature of concentration in the flotation process. Thus, Eq. (4) can be approximated to the following expression.

$$G_i \approx G_j \cdot (1 - Rec) \quad (5)$$

This puts into evidence that the tailings grade originates mainly in the copper grade of the processed ores and in the recovery reached by the process. In this sense, as can be expected based on Eq. (5) for any copper grade found in tailings, there are infinite combinations of feed ore grades and recoveries that can meet the specific tailings grade, so a theoretical concept of Iso-grade of tailings has been developed and is schematically shown in Fig. 3.

In a reprocessing scenario these possible origins of tailings grades could mean different potentials of copper recovery, so not only the copper grade in tailings is important but also the point on the Iso-grade curve indicating historical feed ore grade and recovery. Therefore, a more comprehensive analysis is needed for high copper grades found in old tailings.

2.1.3. Copper speciation in tailings

The Iso-grade concept sustains the fundamental idea that the copper grades measured in any tailings are controlled by the corresponding feed ore grade and recovery, but no quantitative information about the mineralogy of copper in tailings is provided which is the key characteristic that conditions the potential of copper recovery in a reprocessing scenario.

Whether the most suitable recovery process would be flotation or a hydrometallurgical one, the presence of the Non-Sulphide Copper Minerals (NSCM) with respect to the total copper grade requires quantification. Based on Eq. (4) the speciation of copper in terms of the NSCM is expressed as follows:

$$\frac{G_{iNSCM}}{G_{iTOTAL}} = \frac{\frac{G_{cNSCM} * G_{fNSCM} * (1 - Rec_{NSCM})}{G_{cNSCM} - G_{fNSCM} * Rec_{NSCM}}}{\frac{G_{cTOTAL} * G_{fTOTAL} * (1 - Rec_{TOTAL})}{G_{cTOTAL} - G_{fTOTAL} * Rec_{TOTAL}}} \quad (6)$$

But,

$$\frac{G_{cNSCM}}{(G_{cNSCM} - G_{fNSCM} * Rec_{NSCM})} = \frac{G_{cTOTAL}}{(G_{cTOTAL} - G_{fTOTAL} * Rec_{TOTAL})} \quad (7)$$

These expressions must be equal since both represent the same inverse of the mass pull related to tailings.

Then,

$$\frac{G_{cNSCM} * (G_{cTOTAL} - G_{fTOTAL} * Rec_{TOTAL})}{G_{cTOTAL} * (G_{cNSCM} - G_{fNSCM} * Rec_{NSCM})} = 1 \quad (8)$$

And finally,

$$\frac{G_{iNSCM}}{G_{iTOTAL}} = \frac{G_{fNSCM}}{G_{fTOTAL}} * \frac{1 - Rec_{NSCM}}{1 - Rec_{TOTAL}} \quad (9)$$

Eq. (9) called “Speciation Change Equation” represents an expression to calculate the expected speciation of copper in tailings knowing the speciation in the feed ore and the total and specific minerals group recovery reached by the flotation process.

The ratio $\frac{(1 - Rec_{NSCM})}{(1 - Rec_{TOTAL})}$ represents the change in the speciation of copper between the feed ore and the tailings which could be important for scenarios with huge differences between the total recovery and the recovery of a specific mineral or a group of minerals like the NSCM.

2.2. Historical operation data

The following sections summarize the available historical operation data associated to the former concentrator plant that generated the studied old tailings.

2.2.1. Former ore mineralogy

The studied old tailings were originated by a concentrator plant of about 33 million of ore dry metric tons during the 1930’s, averaging a total copper grade of 2.2%. At that time the porphyry copper mine was exploited by underground methods with several points of extraction being in production at the same time. The information from these extraction points puts into evidence the speciation of copper as secondary mineralization, especially chalcocite, which is consistent with the main production zone located in a secondary enrichment alteration.

Although the main copper mineral was chalcocite copper was also found as covellite, chalcopyrite and bornite, as well as oxides, carbonates, silicates, etc., being the latter mineral group not suitable for flotation (Non-Sulphide Copper Minerals-NSCM). According to the historical data about 10% of the total copper contained in the former feed ore was as NSCM.

2.2.2. Former operation data

For the studied site flotation techniques were incorporated to the concentrator plant at the beginning of the 1910’s. Since then the plant underwent several modifications due to operational problems and substantially increased its capacity throughout the years. By the time when the studied old tailings were accumulated the concentrator plant reached a capacity of 18,200 tons per day and comprised the following stages: crushing, grinding, flotation, tailings storage and smelting.

Since the installation of the flotation technology there have been no fundamental changes of how copper is recovered in the plant from sulphide copper minerals. However, considerable improvement is related to equipment and the overall performance of the plant, such as: Semiautogenous (SAG) mill, hydrocyclones, control and automation techniques and new parameters that enable a safer and more controlled operation with a better performance for ores that have become more complex over time, but without modifying the basic processing stages of crushing, grinding and flotation.

Detailed operational data are available for the period when the studied old tailings were generated. Table 1 summarizes the main metallurgical data of the concentrator plant.

A more complete chemical analysis of the different fluxes from the concentrator plant is available from a sampling campaign. Based on this information it has been possible to estimate the recovery of sulphur

Table 1
Operation data of the concentrator plant.

Average ore grade (%CuT)	Average concentrate grade (%CuT)	Average tailings grade (%CuT)	Calculated recovery (%)	Calculated mass pull (%)
2.2	27.1	0.33	86.4	7.1

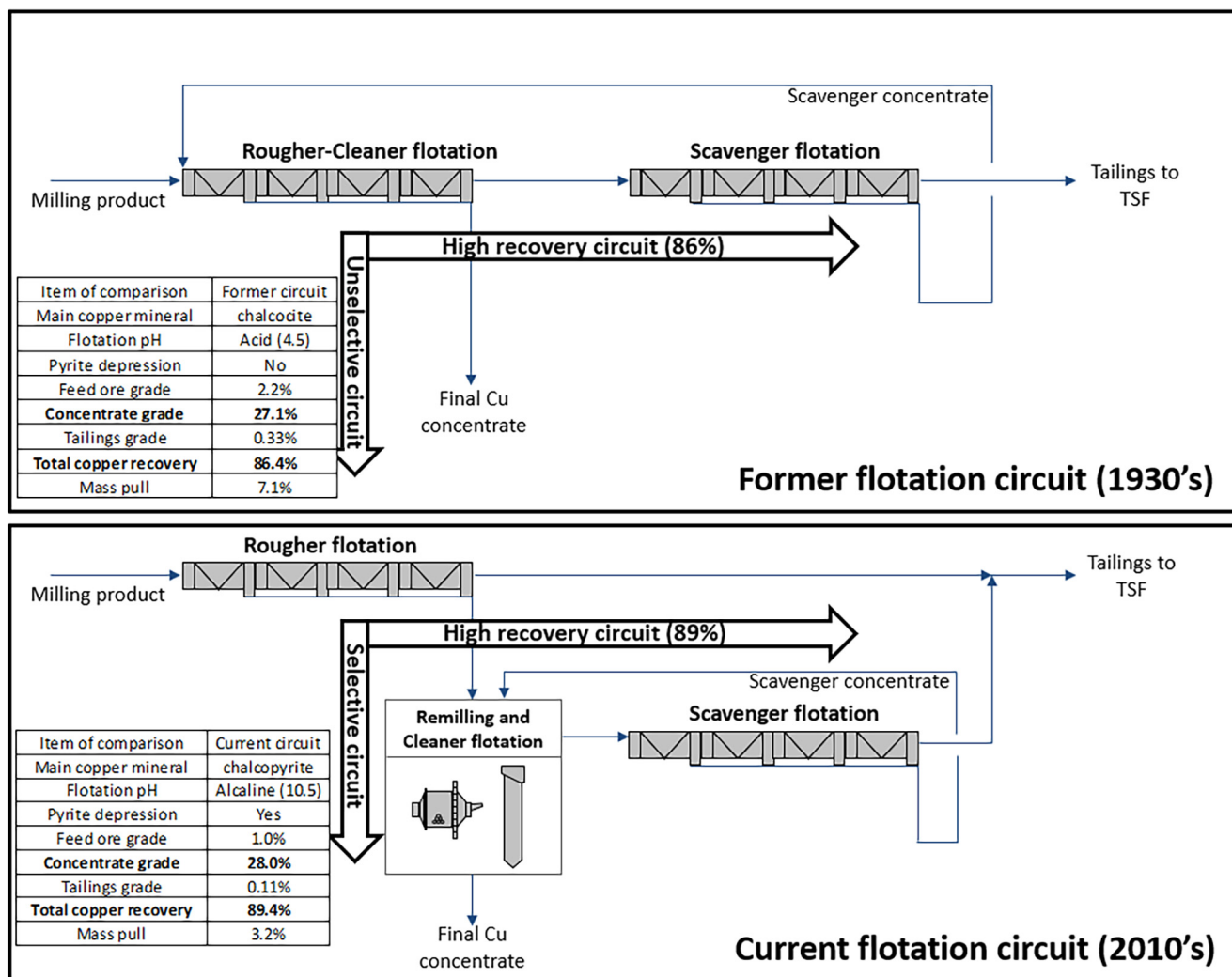


Fig. 4. Comparison between the former and current flotation circuits.

related to the final concentrate as 89.3%. This confirms the fact that during those times the pyrite was floated and not depressed, mainly due to a high degree of liberation after grinding with a target size P_{80} of about 110 μm without regrinding and a flotation pH of 4.5.

3. Results and discussion

In order to make a comprehensive interpretation of the historical operation data, and given the fact that the study case deposit is still under exploitation and operation, a comparison between the former and current flotation circuit including the corresponding main operation data is shown in Fig. 4.

Regarding total copper recovery and concentrate grade the scenario remained unchanged in the past eighty years, a surprising and relevant result considering that the configuration of the flotation circuit, its operational conditions and the ore mineralogy changed. Therefore, other variables of the process should be analysed in detail based on the theoretical concepts already mentioned. In the case of tailings its copper grade changed from 0.33% in the 1930's to current 0.11%. This change occurred practically at a constant recovery being the changes in the feed ore grade responsible for this drop (Iso-grade concept). This is a simple and relevant fact implying for this study case that there is no connection between a supposed inefficient past technology of flotation in terms of recovery with the relative high copper grades found in these old tailings. This study case is not an exception and according to

Gordon (2002) the worldwide recoveries since the very beginning of the flotation technology in the earliest 1920's have not changed much to date.

Taking into account the Speciation Change Equation (Eq. (9)) and considering an optimistic past copper recovery of 30% in the case of the NSCM, then it can be expected that at least 50% of the copper contained in these old tailings has been as NSCM when discharging these materials into the corresponding TSF. At present this percentage could be higher due to the action of possible chemical reorganization processes (Dold and Fontboté, 2001; Johnson and Hallberg, 2005; Akcil and Koldas, 2006) during the past eighty years, a fact that only increases the amount of copper unrecoverable by flotation. Here the Speciation Change Equation clearly shows that a small amount of NSCM present in the feed ore can easily end with a high predominance of these copper minerals in the corresponding tailings. This fact should be kept in mind when measuring and explaining the speciation of copper found in old tailings.

As it can be seen in Fig. 4 the flotation circuit has evolved in the last eighty years, but these changes have been focused mainly on improving the selectivity of the circuit without sacrificing recovery, since more complex ores have been processed along the years. Whereas in the 1930's the main copper mineral present in the processed ores was chalcocite at present it is chalcopyrite with its lower copper content. Accordingly, in the 1930's the selectivity of this flotation circuit was not a relevant task making it easier to reach higher recoveries, as a less

clean final concentrate could easily comply with the copper content that the smelting process demanded; the high mass pull of 7.1% and the not depressing of pyrite is proof of that fact. For the present-day case, chalcopyrite copper content conditions the maximum copper concentrate grade achievable by the process which is already close to a minimum value required for the smelting process implying much effort on selectivity.

For the study case the changes in the selectivity of the flotation circuit and in the mineralogy of the processed ores have occurred simultaneously, annulling their individual effects on the concentrate grade. The inevitable changes in the ore mineralogy with deepening of exploitation levels have been the motor for the changes in the flotation circuit which have evolved to maintain an optimal metallurgical performance of the processed ores in terms of the concentrate grade and without sacrificing recovery.

Findings from this work imply a low copper recovery potential in a reprocessing scenario by flotation, thus new approaches may be more effective in recovering the copper from the studied old tailings such as those ones studied by Falagan et al. (2016). Nevertheless, it should be pointed out that the hydrometallurgical option should deal with the fact that tailings are milled materials limiting its application. For instance, it is not technically possible to build a leach pad with tailings and the leaching in tanks is an alternative which involves high investment and operating costs in solids/liquids separation, an alternative that may be feasibly from a technical point of view but difficult from an economic perspective.

Although this work is based on just one study case its main outcomes may be applied elsewhere, particularly for Chile where several giant porphyry copper deposits are under exploitation since many decades with concomitant generation of old tailings, making this study case highly relevant for the Chilean mining industry, and hence, for worldwide copper mining.

4. Conclusions

An assessment based on historic operation data can provide valuable insights concerning the potential for metal recovery from old tailings in a reprocessing scenario. This step should be the starting point when this kind of information is available. Much effort and costs in analyses and experimental work can be saved with an appropriate interpretation of the historical background.

The historical data from this study case show that flotation was completely optimal in terms of recovery of copper sulphides, favoured by the former less restrictive high copper sulphide mineralogy of the processed ores. Consequently, the main advances in the flotation circuit through the 20th century have focused on improving selectivity without sacrificing recovery due to changes in the ore mineralogy. Accordingly, the main reason behind the relative higher copper grades found in the studied old tailings is just the higher grades of the processed ores, which

is a very relevant finding for the copper recovery potential. Current conventional flotation is not performing better than in the past in terms of recovery, and the higher copper grades of these old tailings probably imply just more copper unrecoverable by flotation.

Acknowledgements

This study has been sparked off by the SecMinStratEl project headed by the TU Bergakademie Freiberg, Germany, and it is part of the corresponding doctoral thesis of the author. Conceptual discussions of the presented ideas with Drs. Nils Hoth, Frank Haubrich (TUBA Freiberg) and Mansour Edraki (SMI, University of Queensland, Australia) are acknowledged. Both comments and suggestions of two reviewers are appreciated.

References

- Akcil, A., Koldas, S., 2006. Acid mine drainage, AMD.: causes, treatment and case studies. *J. Cleaner Prod.* 14, 1139–1145.
- Bugnoson, E., 2001. Country case study on artisanal and small-scale mining: Philippines. *MMSD*.
- Dold, B., Fondbote, L., 2001. Element cycling and secondary mineralogy in porphyry copper tailings as a function of climate, primary mineralogy, and mineral processing. *J. Geochem. Explor.* 77, 3–55.
- Dold, B., Weibel, L., 2013. Biogeometallurgical pre-mining characterization of ore deposits: an approach to increase sustainability in the mining process. *Environ. Sci. Pollut. Res.* 20, 7777–7786.
- Edraki, M., Baumgartl, T., Manlapig, E., Bradshaw, D., Franks, D.M., Moran, C.J., 2014. Designing mine tailings for better environmental, social and economic outcomes: a review of alternative approaches. *J. Cleaner Prod.* 1e10.
- EPA, 1993. Tailings Reprocessing: Magma Copper Company's Pinto Valley Facility. Environmental Protection Agency, U.S.
- Falagán, C., Grail, B.M., Johnson, D.B., 2017. New approaches for extracting and recovering metals from mine tailings. *Miner. Eng.*
- Gordon, R.B., 2002. Production residues in copper technological cycles. *Resour. Conserv. Recycl.* 36, 87e106.
- Graedel, T.E., van Beers, D., Bertram, M., Fuse, K., Gordon, R.B., Gritsinin, A., Kapur, A., Klee, R., Lifset, R., Memon, L., Rechberger, H., Spataro, S., Vexler, D., 2004. The multilevel cycle of anthropogenic copper. *Environ. Sci. Technol.* 38 1253e1261.
- Hansen, H.K., Yianatos, J.B., Ottosen, L.M., 2005. Speciation and leachability of copper in mine tailings from porphyry copper mining: influence of particle size. *Chemosphere* 60 1497e1503.
- Johnson, D.B., Hallberg, K.B., 2005. Acid mine drainage remediation options: a review. *Sci. Total Environ.* 338, 3–14.
- Johansson, N., Krook, J., Eklund, M., Berglund, B., 2013. An integrated review of concepts and initiatives for mining the technosphere: towards a new taxonomy. *J. Cleaner Prod.* 55 35e44.
- Karlberg, A.K., 2010. LKAB: s Skrothögar Är Värda Miljarder/LKAB's Scrap heaps are worth billions. *Ny teknik* 09 (11), 10.
- Kitobo, W., Ilunga, A., Frenay, J., Gaydardzhiev, S., Bast, D., 1999. Bacterial leaching of complex sulphides from mine tailings altered by acid drainage. In: *Hydrocopper 2009*, Antofagasta, Chile, pp. 365e373.
- Lutandula, M.S., Maloba, B., 2013. Recovery of cobalt and copper through reprocessing of tailings from flotation of oxidised ores. *J. Environ. Chem. Eng.* 1 (1), 1085–1090.
- Neethling, S.J., Cilliers, J.J., 2012. Grade-recovery curves: A new approach for analysis of and predicting from plant data. *Miner. Eng.* 36–38, 105–110.
- Rampacek, K., 1982. An overview of mining and mineral processing waste as a resource. *Resour. Conserv.* 9 75e86.