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Mineralogical variations with the mining depth in the Congo Copperbelt: technical and environmental challenges in the hydrometallurgical processing of copper and cobalt ores

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Keywords
Copper-cobalt ores; Mineralization changes; Hydrometallurgical processing; Technologies update; Environment safeguarding

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Abstract

For a long time in the Democratic Republic of Congo, the mining of ores for the production of copper cathodes and salts of cobalt was mostly conducted in the near-surface oxidized mineralization of the Congo Copperbelt. An important part of this mineralization is found in geological settings where copper and cobalt mainly exist as mixtures of oxides and sulfides. With the depletion of surface-rich minerals, mining is taking place at depths where sulfide minerals results in poor metal recovery due to their retention in process wastes. These mineral wastes have the propensity to lead to Acid Mine Drainage (AMD) when exposed to rainfall and atmospheric air. Therefore, major changes in the technologies implemented will be needed in order to guarantee higher process efficiencies while endeavoring to uphold environment safeguarding reinforcement. This work aimed to demonstrate the urgency of updating the hydrometallurgical technologies implemented for the processing of copper-cobalt ores with an emphasis put on minimizing the environmental footprint of process wastes. Four copper-cobalt deposits and downstream hydrometallurgical processes were surveyed to get a better understanding of how changes in mineralization occurring in the deposits will influence the processing technologies and practices during the management of process wastes.

Keywords: copper-cobalt ores, mineralization changes, hydrometallurgical processing, technologies update, environment safeguarding

1. Introduction

For a long time, in the Copperbelt of the Congo, the extraction of copper-cobalt ores was carried out in oxidized deposits close to the surface. The ore products were mainly supplied to concentrators or hydrometallurgical plants [1,2]. The concentrators produced commercial-grade oxide and sulfide concentrates that were either processed at local copper hydrometallurgical plants and smelters, or exported to countries like Zambia and China for further treatments [3-11].

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Similarly, hydrometallurgical plants processed the oxidized copper-cobalt ores and produced both copper cathodes and cobalt salts in the form of hydroxides or carbonates [3,5,12–15]. Since 2012, the DRC has positioned itself as one of the major copper producers in Africa [16], with an annual output of more than one million tons achieved in 2017 [17]. The mineral export statistics for 2016 indicate, for example, that the Metal Mining Group (MMG) and Ruashi Mining, two of the main copper hydrometallurgical plants operating in the region of Haut of Katanga and supplied by the Kinsevere and Ruashi mines, produced an estimated 79,337.283 and 32,139.428 tCu cathodes, and 14,138.412 tCo hydroxides. Boss Mining, another important copper hydrometallurgical plant operating in the Lualaba region, located as Haut-Katanga in the former region of Katanga, produced about 27,601.354 tCu cathodes and 1,686.727 tCo hydroxides by processing ore blends from the Menda, Mukondo, Bangwe, South Kabolela, and Safi open pit mines. It is important to note here that with as the mining depth increased, the majority of the mined ore products contain increasing amounts of sulfide copper-cobalt bearing minerals, which are poorly dissolved in the presence of sulfuric acid used as the conventional leaching solvent at copper hydrometallurgical plants [6]. This results in a steep drop in the recovery of valuable metal and an increasing presence of sulfur in process wastes [18].

Among the strategies designed to meet these challenges, some mining companies are either considering using blends of ores from different mineral deposits to supply copper hydrometallurgical plants or producing commercial-grade concentrates. Such practices cannot be regarded as sustainable solutions for guaranteeing the country’s growth through the extraction of copper-cobalt mineral resources. Therefore, any approach for processing solutions for sustainable mining would require making drastic changes to the processing technologies currently implemented if one needs to improve metal recoveries at copper hydrometallurgical plants [19]. This can be done through the overhaul of the current processing circuits while using up-to-date leaching techniques to adapt to the mineralogical variations of the deposits. In addition, there is a need to focus on strengthening environmental protection by minimizing the footprint of copper hydrometallurgical plants. This can be achieved through improved practices in the management of process wastes given the increased content of sulfide minerals, which can cause AMD in the case of mismanagement [18].

This work seeks to demonstrate the urgency of updating the technologies currently used in the copper-cobalt hydrometallurgical plants in the former region of Katanga in response to observed mineralogical changes in feed characteristics, with an emphasis placed on minimizing the environmental footprint of process waste. To this end, the geological settings of four selected copper-cobalt ore deposits (Kinsevere, Ruashi, Mukondo, and Bangwe mines) were described, with a comprehensive analysis of their mining exploration data. This was followed up with the description and discussion of the downstream hydrometallurgical processing at four selected mines (Mining Metal Group, Ruashi Mining, and Boss Mining, and Gécamines), as well the extent of the process wastes’ environmental footprints and the resulting negative effects, such as the formation and spread of AMD.

2. The origin and location of the Cu-Coore deposit in the Congo Copperbelt

The former region of Katanga, recently divided in four new regions (Haut-Katanga, Lualaba, Haut-Lomani, and Tanganyika), is located in the southeastern region of the DRC, and is home to one of the world’s richest stratiform copper deposits [20–22]. These deposits belong to the Central African Copperbelt that stretches over 700 km from Zambia into the DRC and comprises stratiform copper-cobalt mineralization embedded in the Neoproterozoic rocks of the Katangan Supergroup, consisting of a succession of carbonate and detrital rocks extending over about 10 km [23–25]. The Katangan supergroup was deformed during and after the Lufilian orogeny [26]. It is subdivided into three groups from bottom to top - Roan, Nguba, and Kundelungu groups, including the sedimentary breccias [23,24,27,28]. Most of the well-known copper-cobalt deposits are found in the Roan Group and Mine series (e.g. Bangwe, Etoile, Kinsevere, Mukondo and Ruashi) and Dipeta subgroups (e.g. Shituru) [27,29–31]. Cu–Ag or Cu–Zn–Pb deposits also occur in both the Nguba (e.g. Kamo, Kipushi) and Kundelungu (e.g. Dikulushi, Shangoluwe) groups along the Lufilian arc and its foreland [28,30,32–35]. These mineralizations were precipitated during the early to late diagenesis stages and the syn-to post-orogenic stages [20,30,31,36,37]. The dissemination
and zonation of Cu–Co primary sulfides (chalcopyrite, carrollite, and bornite) within the diverse lithology are the main characteristics of the sediment-hosted strata-bound copper deposits in the Katangan Supergroup [36]. Precipitations of supergene minerals, such as malachite, chrysocolla, and heterogenite, are found in the weathered zone (up to 150 m thick) and constitute oxide ore bodies [32,38–41]. The main mining companies that process oxidized ores are Boss (e.g. Bangwe and Mukondo deposits), MMG (Kinsevere deposit), Chemaf (Etoile deposit), Ruashi mine (Ruashi deposit) and Tenke-Fungurume Mining (Dipeta and Kwatebala deposits).

3. Mineralogical variations with mining depth in the Congo Copperbelt

Copper deposits from the Congo Copperbelt contain cobalt which is the main economic coproduct [41–51]. The upper and weathered mineralized zones of the deposits (80–150 m) comprise oxide and carbonate minerals, such as cuprite, chrysocolla, heterogenite, azurite, malachite, and spherocobaltite [39]. As the depth increases (50–200 m), the deposits become increasingly rich in secondary copper sulfide minerals such as chalcocite, covellite, bornite, as well as chalcopyrite in minor proportions. The secondary sulfide and oxidized minerals result from the weathering of the primary copper and cobalt sulfide minerals of copper and cobalt, chalcopyrite, and carrollite, which are mainly mined beyond 400 m in the unweathered zones of the ore body [45]. The oxidized and sulfide mineralized zones are separated by transition zones with variable thicknesses that contain mixtures of sulfide and oxidized sulfate and oxidized copper and cobalt minerals. For example, in the Kipoi central deposit, located at 45 km from the City of Likasi (Haut-Katanga), the transition zone has a thickness of about 50 m [52]. The transition ore zone extends from around 80 to 130 m of thickness in the Safari and Shaba deposits located along the Kapulo fault in the Katangan foreland [53,54]. In the Tenke Fungurume Mining mine deposit, the oxidized zone extends from 80 to 150 m thickness with around 91% malachite. The thickness of the mixed ore zone underlying the oxidized ore zone (pink cobaltoan dolomite, chrysocolla, and chalcocite) ranges from 50 to 200 m and the sulfide-mineralized zone (chalcoite, bornite, carrollite, and chalcopyrite) extends up to a depth of 1900 m [39]. Fig. 1 presents the KGHM Kimpe ore deposit in the region of Haut-Katanga [19].

The Kimpe ore deposit is located in the southwestern part of the Haut-Katanga region in geological settings that are rich in the mixed copper ores [25,55]. This deposit together with those of Kinsenda and Lubembe, currently mined by the Kinsenda Copper Company, belong to the transition zone separating the Congolese and the Zambian facies [25,55]. The copper mineralization goes in with the mining depth with a slope varying between 65 and 70°. The mixed ores are found in the thickness range of 35 m and 60 m. Beyond the depth of 60 m, copper sulfide minerals are prevalent [19].

Changes in mineralization occur naturally in a deposit with depth. It should be noted that in the deposits, the copper-cobalt mineralization is stratiform and composed mainly of disseminated sulfides in the Roan Supergroup, in the form of lenses or scales (±10 m thick) of the mine series. The Roan is composed of two distinguished mineralized bodies separated by the RSC with varying thickness (0–25 m). Changes in the copper mineralization are related to the origin of the mineralized bodies in the deposits and their weathering degrees [56,57]. Thus, vertical zoning can be established from top to bottom in the deposits. This zoning begins with a totally altered upper zone composed of sterile rocks. This zone is followed by another containing heterogenite in a siliceous and clayey gangue, followed underneath by a zone containing malachite and heterogenite in a siliceous and clayey gangue. Then, comes the transition zone composed of malachite, heterogenite, copper silicates, cobalt carbonates, and sulfide minerals (mainly chalcocite) with a dolomitic gangue. The bottom of the transition zone consists of a non-altered zone, composed of copper and cobalt sulfides and located below the water table [57]. In the former region of Katanga, most of the copper ore is extracted from the transition zone. This is confirmed by data from mining exploration in the central and southern parts of the Lufilian arc, at the Bangwe and Mukundo mines (Kakanda) and Kinsevere and Ruashi mines (Lubumbashi).

3.1. Results of mining exploration conducted in four selected copper-cobalt deposits

This piece of work specifically investigates the mineralogical changes with mining depth in Cu–Co deposits from the Congo Copperbelt [58]. Summarized the grade and tonnage of some Cu–Co deposits in the Congo Copperbelt. Their results showed that the Bangwe mine Cu deposit contained approximately 240,000 Mt of ores with 10,320 Mt Cu (4.3%). The Mukondo mine Cu–Co deposit contained 36,255,529 Mt of ores with about 725,111 Mt and 507,577 Mt @ 1.4% Co. Similarly, the Kinsevere Cu–Co deposit consisted of three mega
breccias of the Mines Subgroup, namely Tshifu-mashi, Tshifu and Kinsevere Hill, extending over ~750, 900, and 1100 m in length and 150–200 m in thickness. It contained about 50,215,648 Mt of ores with approximately 2,053,315@ 4.09% Cu and 426 Mt of Co. The Ruashi Cu–Co deposit comprised three open pits (Ruashi I-III) containing 45,843,595 Mt of ores with 1,286,500@ 2.81% Cu and 112,690@ 0.25% Co. The Etoile Cu–Co deposit, located ~2.5 km to the East of the Ruashi deposit and extended over ~1500 m long, contained 29,697,862 Mt of ores with 1,283,532@ 4.32% Cu and 92,597@ 0.31% Co.

3.1.1. Mining exploration at the Kinsevere deposit

Table 1 presents mining exploration data from 2015 in the Kinsevere deposit. As can be seen, the current mining at Kinsevere consists of over three mega breccias with an anticline mine geomorphology and the average starting level of mining is 1,217 m. The present level of the mining is at an average depth of 1,167 m. The current extraction depth is estimated to be 1,131 m in the transition zone. Mineral exploration at Kinsevere revealed that copper minerals and waste rock contained about 3.5% and 0.5% of the valuable metal. This clearly indicates that the Kinsevere mineralization has changed with depth so the extraction is currently carried out in the deposit transition zone where copper and cobalt are present mainly as oxides and sulfides, as shown by Fig. 2.

At Tshifuia Central mine, the transition zone separating the oxidized and sulfide zones is located between levels 1,160 and 1,070 corresponding to depths of 80 and 130 m, respectively. Currently, the mining of ores is slowed down at MMG Kinsevere and the hydrometallurgical extraction of copper is conducted using concentrates from the deposit of Lupoto where a gravity concentrator (HMS separation) is operated by IVERLAND, a mining company owned by the “Société Minière du Katanga” (SOMIKA).

3.1.2. Mining exploration at the Ruashi deposit

Table 2 presents the mining exploration data of Ruashi deposit conducted in 2015. As shown in Fig. 3, the deposit comprises three ore bodies with a reversed syncline mine geomorphology and the average starting level of the mining is estimated to be 1,282.65 m. The current level of mining is at an average depth of 1,192.56 m with 1,186.67 m corresponding to the depth achieved in the transition zone of the deposit. Mining operations are taking place in the transition zone wherein copper and cobalt occur as mixtures of sulfides and oxides due to changes in the mineralogy of the deposit arising with the depth. The mined ore is treated by the hydrometallurgical route using sulfuric acid.

At present the Ruashi mine is undergoing care and maintenance and the hydrometallurgical plant is fed with ores from other deposits.

3.1.3. Mining exploration at the Mukondo deposit

Table 3 presents the data from the mining exploration conducted at Mukondo. At the Mukondo deposit (Fig. 4), mining operations are conducted in three zones with an average slope of 41° and a starting level of 1,545 m. The current level of mining is on average 1,330 m with 1,320 m as the average depth reached in the transition zone of the deposit where the main copper and cobalt bearing minerals exist both as sulfides and as oxides. Currently, mining is also taking place in the transition zone and as a result, the ROM ores, formerly composed of copper and cobalt oxides, used as feed for the Luita copper hydrometallurgical plant, have
progressively undergone changes in their mineralogical characteristics. For this reason, the mining operator opted for the production of commercial grade sulfide concentrates. At present, the Mukondo mine and the hydrometallurgical plant of Luita are not operational. They are both undergoing care and maintenance.

3.1.4. Mining exploration at the Bangwe deposit

Information related to the mining exploration conducted at Bangwe using data is provided in Table 4.

The mine geomorphology at Bangwe (Fig. 5) is monocline with an average slope of 65° and 1420 m is the starting level of the mining. The present level of the mining averages 1322.5 m and 1267.5 m was the depth achieved in the mixed ores of copper and cobalt. It can be concluded that the mining operations are conducted in the transition zone of the deposit composed of mixed ores with copper occurring both as pseudomalachite and chalcopyrite. Going forward, the mining conducted at Bangwe will lead to the extraction of copper mainly as chalcopyrite, which is a primary sulfide very

Table 1. Data on the mining exploration at the Kinsevere deposit.

| Parameters                                      | Kinsevere Deposit |
|-------------------------------------------------|-------------------|
|                                                 | Kinsevere Hills   | Tshifufia Central | Tshifufia Mashi |
| Mine geomorphology                              | Anticline         | 66°              | 71°             |
| Average slope towards the bottom                | 87°               | 66°              | 71°             |
| Mined levels (m)                                | Starting level    | 1,225.00         | 1,220.00        | 1,205.00        |
|                                                | Present level     | 1,222.5          | 1,110.00        | 1,167.00        |
| Depth achieved in the mixed ores (m)            | ±1,115            | ±1,120           | ±1,120          |
| Mineralized rocks’ types                        | All the mine series except for the RSC |
| Main bearing minerals of Cu and Co              | Oxides: malachite, cuprite, azurite, chrysocolla, heterogenite; |
|                                                | Sulfides: chalcopyrite, bornite, chalcocite, carrollite. |
| Copper average grades in ores and barren rocks  | +3.5% Cu and ±0.5% Cu |
| Cu and cobalt metals reserves                   | 1 390 500 tones Cu and 80 000 tones Co |

Fig. 2. Geological cross-section at the Kinsevere Cu—Co ore deposits: evidence of the boundary of the deposit and the ore — type zone Tshifufia Central Mine Subgroup megabreccias
difficult to leach under the operating conditions ordinarily used at copper hydrometallurgical plants. Consequently, chalcopyrite will remain unleached in the process wastes making them more reactive towards water and air resulting in the formation and spread of AMD. The data from the mining exploration clearly shows changes in mineralization occurring at Bangwe with depth. It is important signaling that mining activities are presently shut down at the open pit of Bangwe.

The analysis of data from the mining exploration conducted at the Kinsevere and Ruashi mines in the Haut-Katanga region and Mukondo and Bangwe mines in the Lualaba region led to the following findings:

- Ore deposits generally present a slope varying between 40° and 70°;
- Mixed copper and cobalt ores are located in the transition zone and in most cases extend beyond a depth of at least 30 m from the surface;
- The amount of sulfide minerals in the ores of a given deposit increases with depth;
- Mining is increasingly carried out in deeper layers of deposits where copper-cobalt mineralization is composed of mixed ores.

4. Processing practices at operational copper-cobalt hydrometallurgical plants

Oxidized ores with malachite and heterogenite as the main bearing minerals of copper and cobalt are first subjected to crushing followed by wet grinding in view obtaining particles with the size smaller that 75–1120 microns. The pulp obtained is sent for sulfuric acid leaching in the presence of sodium
metabisulfite or for paddling of sulfur dioxide which are the reducing agents of the cobaltic cobalt in heterogenite. This is followed by solid-liquid separation in thickeners with the overflow made of high-grade leach liquor directed to clarification prior to purification by solvent extraction (SX). During the purification process, copper is selectively recovered and transferred by means of molecules of oximes, such as LIX 984N or ACORGA OPT 5510, from the leach liquor to the organic phase prepared using kerosene. Afterwards, copper is stripped from the organic phase using the spent from the electrowinning (EW) section prior to its recuperation in the form of cathodes. Also, the underflow composed of solids from the thickener is subjected to washing using counter current decanters and water recovered from the tailing pond. This operation yields low-grade leach liquor that is subjected to purification through the removal of iron, aluminum, and manganese prior to the precipitation of cobalt at approximately pH 8 in the form of hydroxides or carbonates using magnesia or sodium carbonate, respectively. The processing practices in copper-cobalt hydrometallurgical plants in the former region of Katanga consist of the sulfuric acid leaching under reducing conditions, the leach liquor purification by SX, EW of copper and the precipitation of cobalt as hydroxides or carbonates. The processing practices in copper-cobalt hydrometallurgical plants in the former Katanga region are illustrated in Fig. 6.

4.1. Processing practices of ores at the Gécamines’ hydrometallurgical plants

Gécamines is the biggest state-owned mining company in the country and has produced over 450,000 tons of copper between 1970 and 1980. It

Table 3. Data on the mining exploration conducted at the deposit of Mukondo

| Elements of interest                          | Mukondo deposit |
|----------------------------------------------|-----------------|
| Mine geomorphology                           | Anticline       |
| Average slope towards the bottom             | 41°             |
| Mined levels (m)                             |                 |
| Starting level (m)                           | 1,545           |
| Present level (m)                            | 1,325 1,365 1,300 |
| Achieved depth in the mixed Cu–Co ores (m)   | 1,325 1,330 1,305 |
| Mineralized rocks’ types                     | Whole mine series(from RSF to CMN) |
| Main bearing minerals of copper and cobalt   | Oxides: malachite, chrysocolla, heterogenite; Sulfides: chalcocyprite, bornite, chalcocite, carrollite. |
| Average copper grades in ores and the barren rocks | ±2.5% Cu and 0.8% Cu |
| Metal reserves of Cu and Co                  | 598,210 tones Cu and 427,815 tones Co |

Fig. 4. Overview of the Mukondo deposit.
owns the Shituru hydrometallurgical plant which is the oldest plant in the country and uses technology form 1920. Operations started in 1929 and the plant produced annually approximately 30,000 tons of copper cathodes until 1930. Its hydrometallurgical circuits were extended many times and overhauled with the aim of increasing its handling capacity. In 2016, the Shituru plant produced approximately 8,783,181 tons of copper cathodes. This figure is low compared to the annual production of other local hydrometallurgical plants such as Metals Mining Group, Ruashi Mining, Mutanda mine, and Boss mine with 79,337.282; 32,139.428; 191 255.112; and 27,601.354 tons of copper cathodes, respectively. In 2017, the production of the Shituru plant reached 11,399.998 tons which is low compared to 96,735.933 and 25,507.870 tons of Metals Mining Group and Ruashi Mining, respectively. The gap in production between Gécamines and other local plants is very concerning and is mainly due to the use of obsolete technologies, depletion of near-surface oxidized ores, high levels of sulfide minerals in ore deposits and the increasing mineralogical variations with the increased mining depth. The Shituru hydrometallurgical plant has recently been refurbished and the processing technology updated in order to produce copper cathodes using a SX-EW process, as have the majority of plants newly built by the Gécamines in partnership with private investors [59]. Fig. 7 presents a generic flowsheet of the copper-cobalt hydrometallurgical processing used for many decades at Gécamines.

Table 4. Data on the mining exploration at the Bangwe deposit.

| Elements of interest                                      | Makondo Deposit |
|-----------------------------------------------------------|-----------------|
| Mine geomorphology                                        |                 |
| Average slope towards the bottom                          |                 |
| Mined levels (m)                                          |                 |
| Achieved depth in the mixed Cu–Co ores (m)                |                 |
| Mineralized rocks’ types                                  | Monocline       |
| Main bearing minerals of copper and cobalt                |                 |
| Average copper grades in ores and the barren rocks        |                 |
| Metal reserves of Cu and Co                               |                 |

Fig. 5. Overview of Bangwe deposit.
Fig. 6. An SX-EW process used by Mutanda Mining in the former region of Katanga.

Fig. 7. Simplified flowsheet used at copper hydrometallurgical plants operated by the Gécamines.
Gécamines’ hydrometallurgical plants has used for many decades out-of-date processing technologies based on leach liquor purification using the selective precipitation of impurities. This method did not enable high purification levels compared to when solvent extraction is used. In addition, Gécamines used large amounts of different chemicals in order to further purify the leach solutions (Fig. 7). These factors negatively impacted the process efficiency and economic competitiveness. Gécamines produced copper and cobalt metals at its Shituru plant in the Haut-Katanga region, and cobalt carbonate salts at its Kolwezi Copper Plant in the Lualaba region.

4.2. Evolution of practices during the processing of ores at newly built copper hydrometallurgical plants

At the new copper hydrometallurgical plants operated in the former region of Katanga, changes are being made to the processing circuits through the adoption of heap leaching which is a method that is slow in terms of the dissolution kinetics of the bearing minerals of copper and cobalt, but very interesting from an economic standpoint. Indeed, it is not implemented in mechanically or pneumatically agitated reactors requesting electric energy for their functioning. Instead, the heap leaching enables the removal from the SX-EW of the solid-liquid separation stage by thickening after the leaching of ores as well as the washing of the leaching residues using counter current decanters. Additionally, the contact between the ores and the leaching solvent will be lengthened in order to raise the concentrations of the valuable metals to a level enabling the efficient functioning of the process.

Among copper hydrometallurgical plants where Heap leaching (Fig. 8) is common practice in Haut-Katanga, there are those operated by the Société d’Exploitation de Kipoi (SEK) and the Compagnie Minière de Luisha (COMILU), both located almost 75 km from Lubumbashi. These copper hydrometallurgical plants produce copper cathodes using leach liquor from the leaching of ores both in the form of heaps and inside mechanically agitated tanks. Another copper hydrometallurgical plant operated on the same basis is the KAI PENG located along the Kapumpi road leading to the territory of Kambove, which is a mining city located nearly 30 km from Likasi. Gécamines is presently also implementing both the leaching of ores in agitated tanks and in columns and the leach liquor is subjected to purification by SX, with MEXTRAL 984H as the extractant, prior to the electrowinning of copper [59].

In the Lualaba region, Heap leaching of ores is practiced by the copper hydrometallurgical plant operated by Mutanda Mining and recently the one refurbished by the Katanga Copper Corporation (KCC). It is important to note that these two mining companies are owned in a partnership between Gécamines and Glencore International. Heap leaching is also practiced at Boss Mining for the processing of copper oxidized ores.

5. Technical and environmental challenges in the hydrometallurgical processing of copper-cobalt ores

The challenges can be understood in terms of the need to revise the copper-cobalt hydrometallurgical plants processing circuits and to adopt modern and environmentally friendly leaching techniques, such as bioleaching, to adapt to increasing mineralogical changes. Because of the increasing changes in mineralogy, the ore feeds to hydrometallurgical plants are increasingly composed of mixed oxide-sulfide copper-cobalt ores. Due to the current technology being unsuited to this, the sulfide copper-cobalt-bearing minerals are discarded in the process wastes. The presence of sulfide minerals in wastes from the hydrometallurgical processing of ores is a clear indication of the poor dissolution of mixed oxide-sulfide ores when one uses sulfuric acid as a leaching solvent. This often results in low recovery rates of copper and cobalt and high retention of valuable metals in the process wastes as sulfide minerals can lead to AMD in the presence of water and air. Figs. 9 and 10 show the efficiency of the process in selected hydrometallurgical plants.

Boss mining also uses Heap leaching for the processing of the mixed ores of copper and cobalt, which leads to process wastes with sulfur-bearing minerals.

It is clear that the current technologies implemented in hydrometallurgical plants for the processing of copper-cobalt ores have been proven to be ineffective because they cannot adapt to variations in the feed mineralogical characteristics. This calls for an urgent overhauling of the processing circuits at copper hydrometallurgical plants or for up-to-date leaching techniques during the processing of ores with high sulfide minerals contents in order to achieve the highest recoveries of copper and cobalt. Similarly, there is also a need to improve waste management practices in order to minimize their environmental footprint.
Fig. 8. a) Heap leaching of copper ores at the SEK; b) Heap leaching of copper ores at the COMILU; c) Heap leaching of copper ores at the KAI PENG MINING.
In the Congolese Copperbelt, a research has been recently carried out by researchers [60] interested in the leaching of mixed ores of copper and cobalt. They succeeded achieving a simultaneous leaching of copper and cobalt from the ternary system $\text{CuFeS}_2 - \text{Fe}_3\text{O}_4 - \text{Co}_2\text{O}_3$ without the help of traditional oxidizing or reducing agents. Their leaching process is governed by the loop Ferric — Ferrous ions and constitutes an endeavor to be mentioned in direction of the improved dissolution of sulfide-oxide minerals mined in the former region of Katanga. Indeed, after the achievement of variations in the amount of acid, iron oxides and the leaching temperature, the researchers [60] arrived at the conclusion that iron, dissolved in the leach liquor, was liberated by both its oxide minerals and the oxidation of copper from chalcopyrite in the presence of ferric ions. Moreover, the massive presence in the solution of ferric ions has favored both the leaching of chalcopyrite and heterogenite. The latter mineral, through its dissolution, has enabled maintaining the loop ferrous-ferric ions involved in the leaching of oxide-sulfide minerals. In spite of the fact that the studied leaching system...
[60] enables recovering of 89.93% and 95.72% respectively for copper and cobalt, it is evident that its scaling up to an industrial level will require a great number of experiments to ascertain that it will perfectly work when applied to the leaching of mixed ores of copper and cobalt presently mined in Katanga and used as feed to hydrometallurgical plants. This leaching technology could enable recovering more copper and cobalt retained in process wastes as sulfides and thus, prevent them from favoring the formation and spreading of ADM when exposed to water and air.

Among the newly developed leaching techniques, bioleaching can be considered as the most promising when it comes to allowing the dissolution of sulfide minerals by using sulfuric acid as a solvent with both the lowest requirement in energy and the smallest environmental footprint when compared to the pyrometallurgical extraction of metals [61,62]. Bioleaching is already used at an industrial scale in countries such as Chile, China, and South Africa where it is enabling the extraction of base metals from lower-grade ore composed of complex sulfides [62,63]. In addition, this technique has been successfully implemented by researchers [64] who succeeded to conduct the leaching of a polymetallic (Copper, cobalt and nickel) concentrate from the Congolese Copperbelt by means of bacteria. However, a large number of leaching tests should be conducted at the laboratory scale to determine the operating conditions that can ensure the highest recoveries of valuables metals and this could lead to the scaling up of process to an industrial level. It should also be noted that the hydrometallurgical processing of ores from the former region of Katanga using bioleaching would require the mining operators to make very large initial investments for the construction of new plants.

In addition, the bacterial leaching of crushed ores or concentrates previously stacked in columns, heaps, or dumps is a slow kinetic process because of the low growth rate of bacteria [62,65,66]. Therefore, the new plants must be equipped with large columns or highly aerated stirred tanks to successfully leach the sulfide minerals and increase copper concentration in the leach liquors to levels comparable to those obtained during the vat leaching of oxide minerals [62]. However, after a while, the bioleaching process will generate acid, which will result in lower plant operating costs, particularly when one utilizes leaching less acid-consuming ores with a siliceous gangue.

In essence, the bioleaching of minerals is carried out with acidic media using microorganisms consisting of mesophilic bacteria (Acidithiobacillus ferrooxidans, Acidithiobacillus thiooxidans, Leptospirillum ferrooxidans) or thermophilic bacteria (Sulfobacillus thermosulfidooxidans, Sulfobacillus Acidophilus) [66,67]. Sulfuric acid is the main solvent used to leach copper oxides (cuprite, malachite, and azurite) and copper sulfides such as chalcocite (Cu$_2$S) at the atmospheric pressure. Copper sulfide minerals such as bornite (Cu$_9$Fe$_2$S$_8$), covellite (CuS), and native copper are slowly dissolved under such conditions. As for chalcopyrite (CuFeS$_2$), it requires the use of drastic conditions to undergo dissolution in the presence of an aqueous solution of sulfuric acid. As a result, the sulfuric leaching of primary sulfides such as chalcopyrite is well achieved using bacteria or under oxidizing conditions. Poor dissolution, which characterizes chalcopyrite during its sulfuric acid leaching under atmospheric pressure, results from the formation of a less reactive layer made either of Jarosites or sulfur on the mineral surface [60]. Thus, the leaching of chalcopyrite is well understood as a two-step process beginning with the removal of iron from the mineral during dissolution, leaving either a metastable and less reactive layer of chalcocite (Cu$_2$S), a Jarosite or sulfur on its surface. Subsequently, the mineral dissolution begins to release copper, resulting in the formation of a layer of metal-deficient sulfides ultimately leading to elemental sulfur. The latter is a hydrophobic chemical species that slows down and prevents the dissolution of chalcopyrite during leaching with sulfuric acid.

The sulfuric acid leaching of copper sulfide is successfully conducted in the presence of iron and bacteria as catalysts, as shown in Equations (1)–(3):

\[
\text{FeS}_2 + 7\text{O}_2 + 2\text{H}_2\text{O} \rightarrow 2\text{Fe}^{2+} + 2\text{SO}_4^{2-} + 2\text{H}_2\text{SO}_4 \quad (1)
\]

\[
2\text{Fe}^{2+} + \frac{1}{2}\text{O}_2 + \text{SO}_4^{2-} + \text{H}_2\text{SO}_4 \rightarrow 2\text{Fe}^{3+} + 3\text{SO}_4^{2-} + \text{H}_2\text{O} \quad (2)
\]

\[
\text{Cu}_2\text{S} + 10\text{Fe}^{3+} + 15\text{SO}_4^{2-} + 4\text{H}_2\text{O} \rightarrow 2\text{Cu}^{2+} + 10\text{Fe}^{2+} + 12\text{SO}_4^{2-} + 4\text{H}_2\text{SO}_4 \quad (3)
\]

Equations (2) and (3) are related to the production of ferrous ions and their conversion into the ferric ions involved in the bioleaching of copper sulfide minerals, as shown in Fig. 11. The bioleaching of sulfide minerals involves two sub-processes comprising the ferric leaching of pyrite to form ferrous iron and sulfate in the solution and the bacterial oxidation of ferrous iron to the ferric form [62,66].

The bacterial action on pyrite can be direct or indirect (a) and leads to the production of sulfuric acid...
with the conversion of ferrous ions to ferric ions (b) that behave as catalysts of the reaction leading to the dissolution of copper sulfide minerals. Thus, during the leaching of ores, iron undergoes a locked-cycle oxidation/reduction process leading to the conversion of ferrous into ferric ions and the inverse, due to the bacterial action on ferrous ions and the involvement of ferric ions as the oxidizing agents enabling the dissolution of sulfide minerals [62].

Although, the sulfuric leaching of sulfide minerals can take place in the presence of iron and oxygen as oxidizing agents (see reaction (4)), the industrial practice demonstrates that the leaching velocity is increased a million times when it is conducted in the presence of bacteria, such as *Thiobacillus ferroxidans*, *Leptospirillum ferroxidans* and *Thiobacillus thiooxidans*, playing the role of catalysts.

![Fig. 11. Bacterial actions during the leaching process of sulfide minerals.](image)

The bacteria utilize carbon and sulfur as sources of energy for their growth and breeding. A supply in nutrients (ammonium ions and phosphates) together with oxygen is required for the best functioning of the bacterial activity. During the sulfuric acid leaching of sulfide ores, the bacterial activity is optimal when one operates under the following conditions, preferably:

- A medium with a pH between 1 and 6, with 2 as the optimal value;
- An environment with the temperature ranging from 5 to 45°C, with 30°C as the optimal value;
- A supply in oxygen through an injection of air in the heaps of sulfide minerals subjected to leaching.

However, it is important highlighting that the bacterial action can be hindered when the reaction medium contains ions of heavy metals, such as mercury, owing to their bactericidal properties.

6. Improvement of practices during the management of process wastes from hydrometallurgical plants

Sulfuric acid leaching of mixed copper ores generates different types of process wastes. They consist mainly of acidic wastewaters and solid residues, as well as precipitates of impurities. Solid residues are usually pumped as pulp to tailing ponds to enable them settling and separating from the water to be returned back to the leaching stage. Solid residues may also be washed using the countercurrent decanters with the aim of recovering the low-grade leach liquor prior to vacuum filtration in order to obtain cakes. Sometimes tailing ponds are not waterproofed with geomembranes to prevent the release of pollutants into soil and groundwater. In addition, stored tailings are exposed to winds and rainfall, resulting in the release of pollutants to the environment as airborne particles and as leachate. With the observed mineralization changes occurring with depth in the majority of copper deposits, one notes an increase in the amount of sulfide minerals present in the ROM ores. These changes lead to variations in the mineralogical characteristics of the process wastes and their biogeochemical behavior resulting in acidification and AMD as the outcome of exposure to water and air [68]. A glance at Fig. 12 which is related to the SEK mine, a mining company located 75 km from the city of Lubumbashi in the Haut-Katanga, enables comprehending the
environmental issues brought about by the weathering of sulfide minerals. Indeed, in the open pit, one can see acidic solutions and pockets (Grayish zones) of sulfides together with the presence of secondary minerals (whitish crusts) on the mine’s flanks. These signs are evidence of the weathering of sulfide minerals or their alteration in the presence of air and water, ultimately resulting in AMD [68,69].

A series of wetting laboratory-scale tests (Fig. 13) was conducted using waste samples from four copper hydrometallurgical plants fed by ores from the deposits of Mukondo, Bangwe, Ruashi and Tshifuia.

The results given by the first series of tests show that the process waste samples studied released water at a neutral pH. During the second series of tests, a decrease in the pH of the rinsing water was observed up to 3.1–3.2, a value that corresponded to the release of acid by reactions involved in the alteration of the sulfide minerals contained in process wastes. During the third series of tests, the pH of water increased to 3.9. The same trend was observed for the rest of the samples with pH increasing to close to 3.5 after the second washing and rinsing. This variation in the pH reveals a process that was consuming acid liberated by the weathering of sulfide minerals and leading to the occurrence of secondary minerals of copper and cobalt. As the number of cycles of washing and rinsing increased, the acidification process induced by the reaction of sulfide minerals became important so that an acidic pH of between 2 and 3 is recorded revealing the possibility of the formation and spreading of the AMD.

AMD is a phenomenon caused by the oxidation of process wastes containing sulfide minerals or with high pyrite contents [68,69]. As can be seen from the reactions in Table 5, AMD occurs due to the release of sulfuric acid generated by the exposure of the sulfide minerals to water and air [68–70]. The oxidation of sulfide minerals can take place either via the path implicating oxygen or ferric ion, as indicated below [70]:

The absence of neutralizing compounds in process wastes (Table 6) is promoting the setup of chemical reactions involved in the formation and spreading of AMD.

Neutralizing compounds are responsible for the consumption of acid and this phenomenon prevents
plagioclase

Table 5. Oxidation paths of sulfide minerals.

| Oxygen path     | Reaction equation                                                                 |
|-----------------|-----------------------------------------------------------------------------------|
| Pyrite:         | FeS + H₂O + 7/2O₂ → Fe²⁺ + 2SO⁴⁻ + 2H⁻                                           |
| Chalcopyrite:   | CuFeS₂ + 2O₂ → Fe³⁺ + Cu⁺ + 2SO⁴⁻                                              |
| Sphalerite:     | ZnS + 2O₂ → Zn²⁺ + SO⁴⁻                                                          |
| Galena:         | PbS + 2O₂ → Pb³⁺ + SO⁴⁻                                                           |
| Arsenopyrite:   | FeAsS + 3.25O₂ + 1.5H₂O → Fe²⁺ + HAsO⁻ + SO⁴⁻ + 2H⁺                             |

| Ferric iron path| Reaction equation                                                                 |
|-----------------|-----------------------------------------------------------------------------------|
| Pyrite:         | FeS + 14Fe³⁺ + 8H₂O → 15Fe²⁺ + 2SiO⁴⁻ + 16H⁻                                     |
| Chalcopyrite:   | CuFeS₂ + 16Fe³⁺ + 8H₂O → 17Fe²⁺ + Cu²⁺ + 2SO⁴⁻ + 16H⁻                           |
| Sphalerite:     | ZnS + 8Fe³⁺ + 4H₂O → 8Fe²⁺ + Zn²⁺ + SO⁴⁻ + 8H⁺                                |
| Galena:         | PbS + 8Fe³⁺ + 4H₂O → 8Fe²⁺ + Pb³⁺ + SO⁴⁻ + 8H⁺                               |

Table 6. Compounds inducing chemical reaction involved in the consumption of acid.

| Neutralizing compound | Reaction leading to the consumption of acid generated during the oxidation of sulfides |
|-----------------------|---------------------------------------------------------------------------------------|
| Calcite               | CaCO₃ + 2H⁺ → Ca²⁺ + CO₂ + H₂O                                                       |
| Chlorite              | (Mg₃Fe₄₋₂Al₂O₁₀(OH)₈ + 16H⁺ → 4.5Mg²⁺ + 0.2Fe³⁺ + 2Al³⁺ + 3SiO₄ + 12H₂O              |
| Plagioclase           | Na₃₋₂Ca₀.₂₅Al₁₋₂₅Si₃₋₁₇O₈ + 5H⁺ → 0.75Na⁺ + 0.25Ca²⁺ + 1.25Al³⁺ + 2.75SiO₄ + 2.5H₂O |
| Potassium feldspar    | K₂Si₃O₈ + H⁺ + 4H₂O → K⁺ + 3H₂SiO₄ + Al(OH)₃                                       |

the formation and spread of AMD during the storage of process wastes. The same compounds are also retained through the precipitation of metal ions contained in the water liberated by the AMD and this process results in the confinement of the pollution that can affect water and soil. However, this is not the case for the majority of process wastes generated by copper hydrometallurgical plants operational in the former region of Katanga, because neutralizing compounds are removed during the leaching of ores by sulfuric acid. When storage areas are not waterproofed or lined, water and soil pollution associated with alterations of sulfide minerals occurs due to reactions with water and air that cannot be avoided.

7. Conclusion

In the former region of Katanga, copper-cobalt ores from open pit mines revealed a significant variability of mineralization which deepens with the depth of mining, with a slope varying between 40 and 70°. Data from the mining exploration collected at four selected copper deposits showed that mining is increasingly taking place in the transition zones which are mainly composed of mixed ores of copper and cobalt. Therefore, large amounts of valuable metals as sulfide minerals are found in ores. Sulfide minerals react poorly to dissolution from sulfuric leaching technology, which is currently implemented in copper hydrometallurgical plants and, consequently, the recoveries of copper and cobalt are barely at a level of 75%. As a result, the content of copper in process wastes can increase by up to 0.7% due to its retention as residual sulfides that can cause AMD during storage. It is clear that there is an urgent need to update the current technologies implemented at hydrometallurgical plants in former Katanga and to reinforce the minimization of the process wastes environmental footprint. To this end, bioleaching techniques are promising as they quell these threats and can at the same time help-enhancing the dissolution of sulfide minerals contained in mixed copper-cobalt ores presently used as feed to copper hydrometallurgical plants. It is believed that implementing bioleaching techniques will increase the recovery of copper beyond 75.0% and delay AMD forming and propagation through the removal of sulfur from process wastes before their storage.

Conflicts of interest
None declared.

Ethical statement
Authors state that the research was conducted according to ethical standards.

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