

# Study of Cu Recovery Comparison of Fresh, Stockpiled, and Mixed Ores Processing in Batu Hijau Mine – Sumbawa Island, Indonesia

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## ABSTRACT

Batu Hijau mine is one of the mining areas in Indonesia that have been operated for 20 years. The main concentrate product in Batu Hijau contains the primary minerals copper (Cu) and gold (Au). However, the mine is not only processing the fresh mined ore but also store the low-grade ore in the stockpile. Therefore, this study compares the Cu recovery in different ore stockpile processing on and changes in the concentration of dissolved Cu in the tailings that potentially contaminated the environment. Chemical analysis of solid and dissolved Cu uses standard methods by strictly carrying out quality assurance and quality control (QA/QC) procedures to ensure the resulting data has high validity. The results showed that there is a slight increase in Cu's solubility in liquid fraction of tailings and the recovery decrease when ores were processed from stockpiles. However, the recovery reduction is anticipated by flotation with controlled potential sulphidisation (CPS) using sodium hydrosulphide (NaHS). Besides, the CPS also ensure that only small amount of dissolved Cu that dumped in the tailings. Hence, the dissolved Cu concentration can be monitories to reduce the possible impact on the environment.

Keywords: Dissolved Cu, Ore, Recovery, Stockpile

## INTRODUCTION

Batu Hijau mine, located on Sumbawa Island, is one of the largest mining sites in Indonesia. The Batu Hijau focused on two main minerals, copper (Cu) and gold (Au). Since 2000, the mine has produced around 3.6 million tonnes of copper and 8 million ounces of gold [1]. Notably, a typical mining industry that produces copper and gold might generate mining waste in the form of dissolve metals such as copper itself, arsenic, chromium, cadmium, and others [2]. To manage their waste, the Batu Hijau utilizes Deep Sea Tailing Placement (DSTP)

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to prevent contaminant exposure to the environment. However, the mine needs to diminish the number of wastes by increasing the targeted metal recovery rate during the processing stage. Thus, an effective processing system is essentially required to reduce the number of heavy metals dumped into the tailing [3].

During the processing phase, the extracted ore from the pit is processed into concentrate using the flotation method with Controlled Potential Sulphidisation (CPS). The CPS is carried out by adding sulphidisation reagents such as Sodium Hydrosulphide (NaHS) or Natrium Sulphide (Na<sub>2</sub>S) to amplify the Cu recovery during the flotation process [3,4]. Still, not all mined ore is immediately processed into concentrate. At the beginning of the mining operation period, only the high-grade ore were directly processed, while the low-grade ore and some medium-grades were stored as stockpiles for later processing [5].

When the ore are stored, some are oxidized by air and water, causing the stockpiled ore to have different properties from the fresh ore. The most noticeable difference is that metals from the oxidized ore become much more soluble in water and form the Acid Soluble Copper (ASCu) on the surface of the ore. This condition might reduce the mineral's floatability and lower the targeted mineral recovery rate [6, 7]. On the other hand, low recovery rate and the existence of ASCu might impose severe treat to the environment. The minerals that cannot properly recovered during the main processing stage will possibly be culminated in the tailing while the ASCu could form the acid main drainage (AMD), a hazardous substance for the environment [2, 3]. In this case, the addition of sulphidisation reagents is highly likely to increase the recovery rate by recreating the sulfidic layer on the surface of the oxidized ore and decreasing the concentration of ASCu [7, 8].

In Batu Hijau, sulphidisation is not the only way to improve the Cu recovery, yet they also vary the ore mixture between the fresh and stockpiled ore. Three main feed combinations have been established: feed dominated by fresh ore, feed dominated by stockpiled ore, and feed consists of only stockpiled ore. The ore combinations produce different recovery rate which in turn affect the percentage of the targeted minerals in the tailing. Hence, this article compares the Cu recovery on the processing of the ore based on its mixture (fresh, stockpiled, and mixed ores) and associate the Cu recovery with the ASCu concentration for 20 years period of time. This study also observes the dissolved Cu in the tailing, to ensure the ore processing is not exceeding the national standard for heavy metal concentration in the tailings, which possible to disturb the environment.

To obtain the intended purpose, data for this study has been collected continuously for 20 years from the early time the mining started to operate in 1999 to 2019. The samples analyzed in the laboratory were obtained from the processing plant of Batu Hijau mining site where ore is processed and the product of concentrate and tailing are produced. All samples were collected following internationally standardized procedure in mineral and wastewater analysis, including American Society for Testing and Materials (ASTM) E2941, International Organization for Standardization (ISO) 10258, American Public Health Association (APHA) 3125B, and the use of Inductively coupled plasma mass spectrometry (ICP-MS).

## EXPERIMENT

### Chemicals and instrumentation

Most chemicals used in this study were analytical grade, and some were spectroscopic grade. Aquabidest, aquadest, perchloric acid (HClO<sub>4</sub>), hydrochloric acid (HCl), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), ammonia (NH<sub>3</sub>) and acetic acid (CH<sub>3</sub>COOH), ammonium hydrogen difluoride,

potassium iodide (KI), sodium thiosulfate ( $\text{Na}_2\text{S}_2\text{O}_3$ ), starch, and nitric acid ( $\text{HNO}_3$ ) were employed.

Instruments applied were Atomic Absorption Spectroscopy (AAS, HITACHI Z-2000 GBC 932 AA) and Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) Agilent 7800.

### Procedure reaction

To achieve the aim of this research, two primary analyses were conducted. Firstly, the analysis of copper concentration in ore, tailing, and concentrates to determine the recovery of copper. In addition, concentration of ASCu as a result of oxidation that might happen in the stored ore (stockpile) was also examined. Hence, it is possible to compare the ore characteristics and Cu recovery as well as anticipating ASCu formation in the processing plant of Batu Hijau Mine. Secondly, analysis of the dissolved copper concentration in tailing to examine the amount of copper dumped into tailing in the liquid state. This analysis was carried out to confirm that the ore processing is not leaving dissolved Cu into the tailing and harming the environment.

The analytical methodologies employed in this study were based on the standardized procedure that has been widely used in mineral and wastewater analysis. The method involves the sampling and sample preparation procedure as well as the main analysis procedure. The list of standard methods used in this study is served in Table 1.

**Table 1.** List of Standard Methods

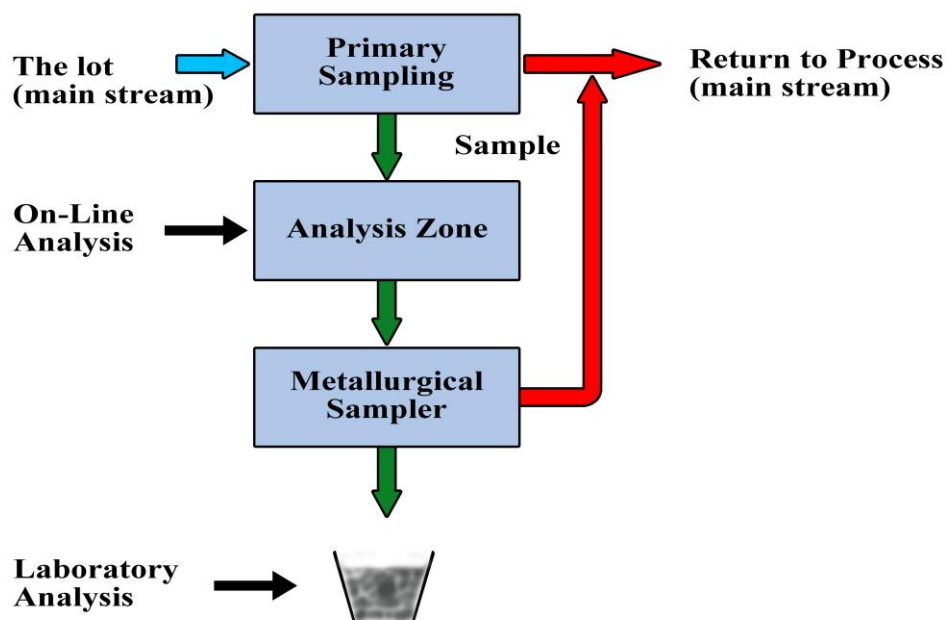
Testing Material	Reference Methods		
	Sampling	Sample Preparation	Testing
Ore, Tailing	ISO 11794	ISO 12743	ASTM E2941
Concentrate	ISO 11794	ISO 12743	ISO 10258
Liquid Tailing	SNI 8520 & SNI 6989.59	SNI 8520 & SNI 6989.59	APHA 3125B, ICPMS

### Cu Concentration Analysis in Ore, Tailings, and Concentrates

The Cu concentration analysis was conducted in three different sampling locations, ore, tailings and concentrates to understand the ore characteristics, to examine the Cu recovered from the concentrates, and to determine the amount of Cu dumped into the tailings. Therefore, the ore characteristics and Cu recovery in production process can be compared. In general, the sampling was done at the main Processing Plant. The ore sample was taken from the cyclone overflow (commonly referred to as a feed). Meanwhile, the tailings samples were collected from the Tailings deaeration box (Tailbox), and the sample of concentrates was taken from the column/cleaner (Figure 1). The sampling procedure at the processing plant for ore, tailings, and concentrate referred to the automatic sampling method under the ISO 11794 (Figure 2) [9].



**Figure 1.** Sampling Locations on Processing Plant for Ore, Tailing, and Concentrates



**Figure 2.** Principles of automatic sampling

Before being tested, the sample was prepared to ensure the compatibility of the sample and the test method. The sample preparation process referred to ISO 12743. The ore, tailings, and concentrate samples were sent to at the QA/QC Lab in the form of slurries. The sample was then drained in a vacuum filter for about 10 minutes or until the slurry transformed into cake form with less water concentration. Subsequently, the cake-form sample was dried in an oven before being grounded and mashed into powder [10]. The samples were then ready to undergo the testing procedure.

Two standard methods were utilized to determine the copper concentrations, namely ASTM E2941 and The ISO 10258. The ASTM E2941 procedure tests low copper concentrations, such as in rock (ore) and tailings. The procedure began with decomposing the sample with 3 ml Perchloric Acid (HClO<sub>4</sub>) and 5 ml Chloride Acid (HCl). The mixture was then heated on a hot plate until dry. After that, 10 ml of HCl, aquabidest, and flocculant were added to the sample. The Cu concentration was measured using an atomic absorption spectrophotometer (AAS) instrument [11].

Acid-soluble copper (ASCu) content in the ore was also determined. This was to confirm whether the oxidation occurred on the ore surface, especially in the stockpiled ore [6]. The ASCu was examined by adding 5% of sulfuric acid to the ore samples. The mixture was then shake in a shaker for 60 minutes and filtered to obtain filtrate and to determine copper calibration solution with 5% of sulfuric acid in the AAS.

Meanwhile, The ISO 10258 titrimetric method was used to analyze samples with high Cu concentrations, such as concentrates. The prepared sample was digested with nitric acid (HNO<sub>3</sub>) on a hotplate. Once the brown fumes disappear, potassium chlorate (KClO<sub>4</sub>) was added. Subsequently, sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and aquabidest were mixed to re-dissolve the solution. To neutralize the solution, Ammonia (NH<sub>3</sub>) and Acetic Acid (CH<sub>3</sub>COOH) were added to the mixture before adding Ammonium hydrogen difluoride. After that, Potassium Iodide (KI) was added, and the mixture was titrated immediately with Natrium thiosulfate (Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>) as standard volumetric solution and starch as an indicator [12].

### **Dissolved Cu Analysis in Tailing**

Tailing samplings were carried out at the deaeration box (tailbox) by a certified field officer with a procedure referring to SNI 8520 and SNI 6989.59. The tailing sample comprised samples taken in the morning, afternoon, and evening on one sampling day. The slurry tailing sample was prepared and deposited in a container (1L) for 15 minutes. After that the liquid and the solid fractions of the slurry sample were separated. The liquid fraction was then transferred to a bottle for filtration (0.45 mm diameter) and preserved using 10% HNO<sub>3</sub> for dissolved copper measurements [13, 14].

The dissolved copper analysis was conducted using the APHA 3125 B method with Inductively Coupled Plasma–Mass Spectrometry (ICP-MS) as a measurement instrument. At the beginning of the analysis, the prepared sample was digested by mixing the samples with an acid preservative, aquadest, and an analyte in a polypropylene tube. After that, HNO<sub>3</sub> was added to samples, blank solution, and standard solution before being heated at 105 °C. During the heating, Nitric acid (HNO<sub>3</sub>) was added until the digestion process completed for 2 hours. The samples were diluted with aquadest to reach 10 ml volume after being cooled. The diluted sample was stored at 4 °C temperature, before the ICP-MS analysis was conducted [15].

## **RESULT AND DISCUSSION**

### **Comparison of Cu Recovery on Ore Mixture Variation**

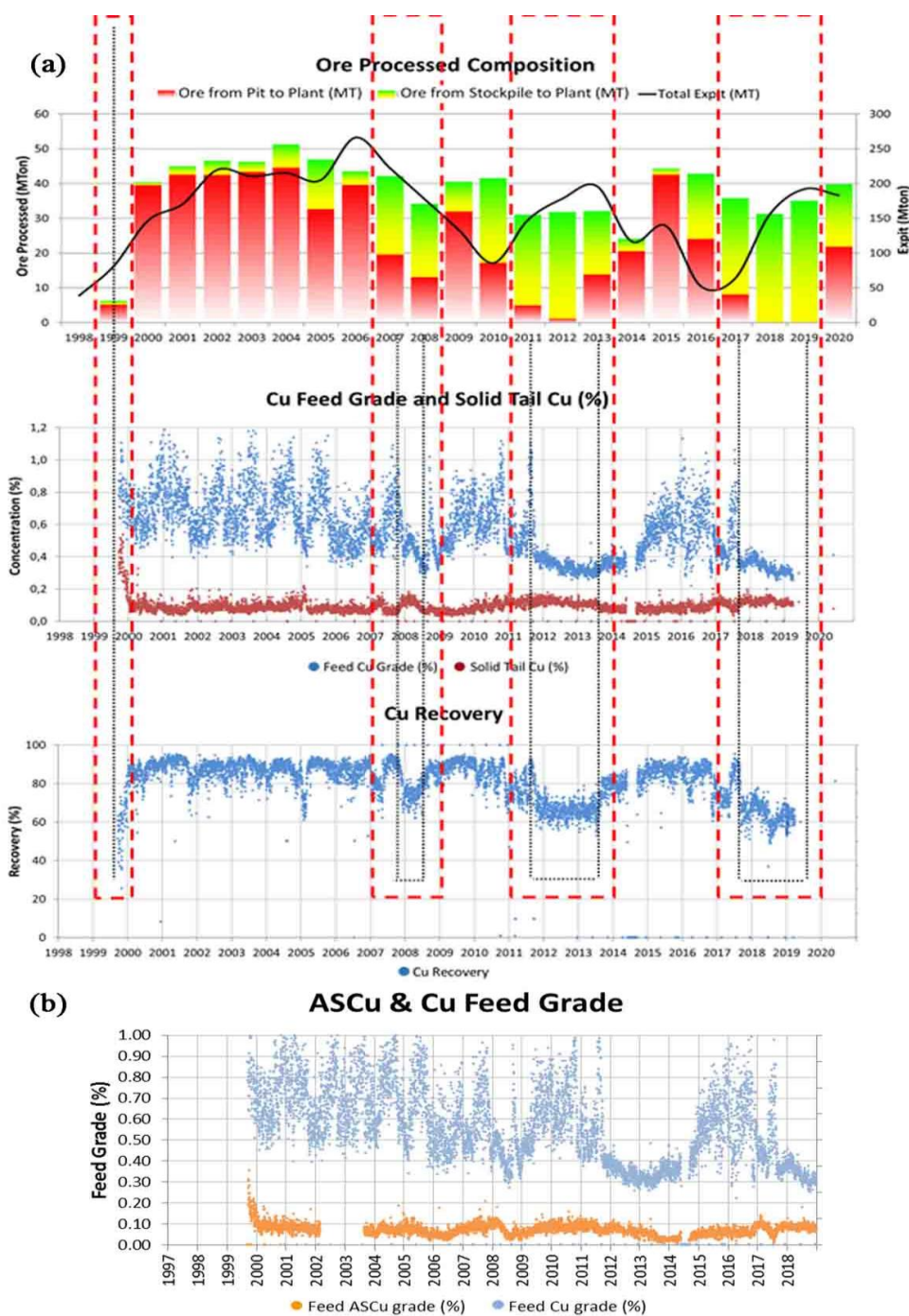
The chemical analysis of copper concentrations in ore and tailings shows that the percentage of Cu recovery in the concentrator plant is inversely related to the number of stockpiled ore being processed (Figure 2(a)). The higher the amount of stockpile processed, the lower the amount of Cu recovery obtained. This trend can be observed in the production periods of 2007-2008, 2011-2013, and 2017-2019 periods when the majority of ore processed was stockpiled ore, as shown in Figure 3(a).



The most significant decrease in recovery percentage can be seen in 2018-2019, when all ore processed was stockpiled ore. In this period, the rate of Cu recovery ranges from 50% - 80%. Similar percentages were also found for other periods where the majority of ore processed was stockpiled. On the other hand, the rate of Cu recovery in periods where the majority of the processed ore was high grade fresh ore, such as in the 2000-2004 period, the percentage of Cu recovery was above 80%. Thus, it can be concluded that the stockpiles mixture in the production process might lower the recovery rate. Meanwhile, higher number of fresh ore in the concentrate can escalate the rate of Cu recovery significantly.

The decrease in Cu recovery is happening since the stockpiled ore undergoes an oxidation reaction with free air. The ore mined at Batu Hijau contains chalcopyrite ( $\text{CuFeS}_2$ ) and bornite ( $\text{Cu}_5\text{FeS}_4$ ) as the main minerals of Cu and various other minerals, such as Pyrite, Plagioclase, quartz, biotite, chlorite, and Sericite. All of these minerals have the potential to undergo oxidation reactions [6]. However, among these minerals and several other sulphide minerals such as Sphalerite and Galena, Pyrite ( $\text{FeS}_2$ ) is the mineral with the highest oxidation rate [16, 17].

Pyrite oxidation can produce sulfuric acid ( $\text{H}_2\text{SO}_4$ ), which can accelerate the dissolution of other metals in the ore, including Copper (Cu), Zinc (Zn), Cobalt (Co), Iron (Fe), and others [6]. However, Cu is known to dissolve much more rapidly in sulfuric acid than other metals [18]. Cu dissolved in sulfuric acid is known as acid-soluble Cu (ASCu). The relationship between the metal dissolving process and the concentration of ASCu formed is shown in Figure 2(b). The data in Figure 3(b) shows that the amount of ASCu is influenced by the number of Cu present in the feed or the ore. The higher the number of Cu in it, the higher the amount of Cu that can dissolve in  $\text{H}_2\text{SO}_4$ , formed due to pyrite oxidation. Cu recovery is associated with the amount of ASCu. According to Figure 3(b), when the Cu recovery decline to below 80% the ASCu percentage tend to rise slightly. This trend can be observed during 2007-2008, 2011, and 2017-2018, where the stockpiled ore dominated the mixture.



**Figure 3.** (a) Effect of feed from stockpile on Cu recovery; (b) ASCu percentage on feed

In addition to producing  $H_2SO_4$ , Pyrite oxidation also produces  $Fe^{3+}$  ions, accelerating chalcopyrite's oxidation [19]. The reaction is:



Besides being oxidized by  $Fe^{3+}$ , Chalcopyrite [20, 21] and also Bornite [22] can undergo direct oxidation with air through the following reactions:



The effect of Pyrite on strengthening Bornite and low-grade Copper ore bioleaching has been reported in previous studies by the presence of microorganisms [23–25]. The oxidation reactions experienced by these two types of sulphide minerals can then affect their surface properties. The oxidized surface of sulphide, chalcopyrite, and bornite minerals can affect the selectivity of separation between valuable sulphide minerals, in this case, Chalcopyrite and Bornite, and Gangue (worthless sulphide minerals, such as pyrite, silica) in the flotation process [17,26]. This happens since the surface of the oxidized minerals generally becomes hydrophilic [27]. In addition to the oxidation reactions caused by free air, sulphide minerals can also undergo oxidation reactions due to interactions between sulphide minerals such as Chalcopyrite and Pyrite.

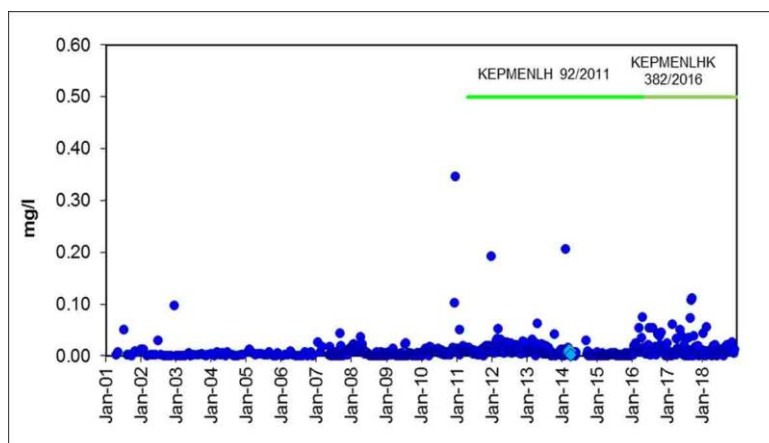
The interaction between these two minerals causes the emergence of Galvanic Interaction, which causes Chalcopyrite oxidation and activates copper on the pyrite surface. Activated copper on the pyrite surface can increase collector adsorption on the pyrite surface. This situation can increase pyrite flotation and, at the same time, reduce chalcopyrite recovery [28,29]. Galvanic interactions also cause the formation of a hydrophilic layer on the surface of iron (II) hydroxide, which acts to inhibit chalcopyrite's flotation [29–31]. As a result, the percentage of chalcopyrite recovery is reduced. The low chalcopyrite recovery in this flotation process will increase the Cu concentration in the tailings. Therefore, it is essential to determine the dissolved Cu concentration in tailings to monitor the possibility of copper to be released to the environment.

### **The Concentration of Dissolved Cu in Tailings**

The presence of ASCu is one of the factors that cause a decrease in copper (Cu) recovery in the flotation process. ASCu will not be recovered in the flotation process and eventually be deposited in the tailings. The data presented in Figure 4 shows dissolved Cu in the liquid fraction of tailings since the mine began operations in 2001. In general, the number of dissolved Cu concentration in the liquid fraction of tailings can be said to be relatively constant well below the Government of Indonesia permitted limit for discharge (KEPMENLHK 92/2011 and 382/2016), although, in specific years, there have been spikes with high concentrations, such as what happened in 2010 (0.346 mg/L), 2012 (0.192 mg/L), and in 2014 (0.206 mg/L).

The presence of ASCu is an indication of oxidation process. Among other types of ore, the stockpiled ore are the ones that get oxidized the most. Generally, oxidized ore produces copper sulfate or copper oxide, depending on the mineral being oxidized. Copper sulfate is soluble in polar solvents such as water. This condition is the reason why copper minerals become dissolved. In other words, the more copper sulfate that is formed due to the oxidation process, the greater the possibility of finding dissolved copper concentrations in the liquid fraction of tailings.





**Figure 4.** Dissolved Cu concentration in liquid fraction in tailings

The data in Figure 4 show that when more stockpiles were used in the production process in 2007-2008, 2011-2013, and 2017-2018, the dissolved Cu concentration in the liquid fraction of tailings was not significantly affected though there was an increase. However, this increase is of small value and is always below the Indonesian government's standard. For example, the increase in 2008 was only 0.018 mg/L. The highest concentration of dissolved copper was measured in 2011, 0.346 mg/L, followed by a concentration of 0.206 mg/L in 2014. Both values are still below the national standard of Indonesia. The results indicate that the mining process is capable to prevent the excessive amount of dissolved Cu through the Controlled Potential Sulphidisation (CPS) technique in the flotation process.

The CPS technique can maximize the recovery process during flotation, for the amount of NaHS used will depend on the feed variation. The addition of NaHS, a sulphidising agent, can restore minerals' ability to stick to air bubbles during the flotation process. This process can occur because the mineral surface that has been oxidized can adsorb hydrosulphide ions originating from NaHS to form a sulphide layer on the mineral surface covered with iron copper oxide/hydroxide [32]. The sulphidisation mechanism of oxidized chalcopyrite is known to parallel the sulphidisation mechanism of malachite:



However, the addition of NaHS must be at the right level because adding too much or too little can reduce the percentage of Cu recovery during flotation. Higher recovery will result in less amount of Cu ending up in the tailings [8].

## CONCLUSION

The measurement results for twenty years show that incorporating stockpiled ore during production process causes Cu recovery to be lower (50-80%) when compared to the recovery of the production process using only fresh ore (> 80%). Another consequence of the stockpiled ore usage is the increase in the concentration of dissolved Cu in the liquid tailings. This occurred when the stockpiled ore dominate the mixture. This growth occurred due to ore's oxidation reaction, indicated by the rise in acid-soluble copper's value, which was higher than its value in fresh ore dominant mixture. However, the increase in copper dissolution that occurs can be anticipated by the application of NaHS reagent. Hence, even though the Cu recovery rate might

vary throughout the 20 years of production, the dissolved Cu ended up in the tailings is not exceeding the standard by Indonesian Ministry of Environment and Forestry.

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