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EFFECT OF THIOSULFATE CONCENTRATION AND LEACHING TEMPERATURE IN AMMONIACAL THIOSULFATE LEACHING OF REFRACTORY SULFIDE GOLD ORE

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ABSTRACT

Thiosulfate as gold leach solution first studied in 1979 to found alternative of cyanide and mercury solution which are widely used in gold extraction industry although it's negative impact for the environment. The ore sample is native ore from Bolaang Mongondow, North Sulawesi. According to Optical Microscope observation and LIBS characterization, there are pyrite compound which is one of the chacaracteristic of sulfide ore. According to X-ray fluorescence and Inductively Coupled Plasma, the ore contained about 14.62% Fe, 6.69% S, 0.15% Cu, and 0.27 ppm Au. This study aimed to determine the effect of thiosulfate concentration and leaching temperature in ammoniacal thiosulfate leaching of refractory gold sulfide ores on the solubility of gold. This research was conducted by laboratory scale of leaching method. The leaching result is then checked by Inductively Coupled Plasma (ICP). The concentration of thiosulfate (0.05M, 0.1M, and 0.2M) and the effect of temperature (25^0 C, 40^0 C, 60^0 C) were studied. Maximum gold extraction (62%) was obtained using 0.1M of thiosulfate concentrations at 40^0 C for 2 h with 20% of pulp density. Stirring speed and the pH of the aqueous solution were 400 rpm and 10, respectively. According to the results, it can be concluded that the concentration of Thiosulfate solution and leaching temperature will affect the dissolution of gold.

***Keywords**: Bolaang Mongondow; Non-Cyanide Leaching; Refractory Gold Ore; Thiosulfate Leaching*

INTRODUCTION

Bolaang Mongondow is a region in North Sulawesi, Indonesia, containing gold from medium to high-grade sulfide mineralization (Kavalieris et al., 1992). Due to the effectiveness and high selectivity of gold in the gold dissolving process, cyanide and mercury

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have been employed as reagents for the extraction of gold from mineral sources (Asamoah et al., 2014; Marsden & House, 2006) since it was first introduced by John Stewart MacArthur in the 1880s (Hilson & Monhemius, 2006).

Despite this, there are growing health and environmental concerns regarding its use, notably in Indonesia, due to its detrimental health effects. a contaminated, disease- and death-causing environment (Rice et al., 2014; U.S. Department Of Health and Human Services, 2002). In addition, cyanide leakage from metallurgical companies have resulted in several significant environmental incidents throughout the world (Hilson & Monhemius, 2006; Jiang, 1998; Muir, 2011). As a result of environmental concerns, cyanide leaching is now prohibited in many regions. In addition, the typical cyanidation leaching time is 24 hours (Jiang, 1998), and gold cannot be successfully extracted from refractory gold ore. Consequently, alternate lixiviants to gold have gained increasing interest in recent years. The most attention has been paid to chloride, thiourea, and thiosulfate among these lixiviants.

Berezowski and Sefton began conducting intensive research on Thiosulfate as a reagent for gold leaching in 1979. Following this, the effect of leaching solution concentration, pH, and leaching process temperature on the dissolution of gold from different types of ores from different countries was examined using a variety of ore types from various countries. Thiosulfate leaching is widely regarded as the most promising alternative technique due to its acceptable gold leaching rate, environmental friendliness, high reaction selectivity, low corrosivity of leaching solutions, inexpensive reagents, and ability to leach high-grade gold ores containing copper and carbon materials, etc (Feng & van Deventer, 2011; Jeffrey et al., 2003; Lampinen et al., 2015; Senanayake et al., 2011; Xu, Yang, Li, Jiang, et al., 2016; Xu, Yang, Li, Yin, et al., 2016).

In recent years, the copper-ammonia-thiosulfate leaching system has garnered significantly more interest. Numerous studies have been conducted on thiosulfate leaching systems in terms of thermodynamics, leaching kinetics, speciation distribution, as well as the stability of thiosulfate and diverse gold recovery strategies. In Ammoniacal Thiosulfate leaching, there are main reagents for the process, Thiosulfate, Ammonia and Copper (II) Ions. Thiosulfate is the main reagent in this process because it will form thiosulfate-gold complex with gold (Jeffrey, 2001). Copper (II) ions becomes a catalyst that can increase leaching rate by up to 20 times (Gradov, 2013). Nevertheless, copper ion has negative effect, it can accelerate thiosulfate degradation and increase thiosulfate consumption during the process. This makes it necessary to add an additive to stabilize copper thiosulfate solutions. Ammonia has been used as an additive to stabilize copper ion in this process. Moreover, ammonia prevents the thiosulfate decomposition on the surface of gold which leads to the gold passivation (Oraby, 2009). The possible mechanisms for gold leaching using thiosulfate solution are as follows (Mohammadi et al., 2017):

$$2Au + 10S_2O_3^{2-} + 2Cu(NH_3)_4^{2+} \rightarrow 2Au(S_2O_3)_2^{3-} + 8NH_3 + 2Cu(S_2O_3)_3^{5-} \quad (1)$$

$$4Cu(S_2O_3)_3^{5-} + 16NH_3 + O_2 + H_2O \rightarrow 4Cu(NH_3)_4^{2+} + 4OH^- + 12S_2O_3^{2-} \quad (2)$$

Copper (ii) ions-ammoniacal-thiosulfate leaching of Bolaang Mongondow gold ore has not been comprehensively discussed in other studies, according to the author's best knowledge. This study aimed to determine the effect of thiosulfate concentration and leaching temperature in ammoniacal thiosulfate leaching of refractory sulfide gold ore from Bolaang Mongondow, North Sulawesi, Indonesia.

METHODS

The gold ore sample was obtained from Bolaang Mongondow, North Sulawesi, Indonesia. The ore was crushed by a hammer until the ore size can be processed using a ball mill machine. The ore was milled by a planetary ball mill (Puluerisette 6), the milled ore will be sifted with sieve machine (RETSCH AS200) with strainer size 80 mesh. After that, a sample be roasted using carbolite furnace at 500 degrees celsius for 2 hours. The roasted sample will be milled and sieved with a size of 200 mesh for the leaching process.

For initial characterization, we use aqua regia test. 5 gram of sample was used. We used combination of HCl (13 M, Merck) and HNO3 with ratio 3:1 as reagent. The solution of reagent and sample were carried out in 100 mL Erlenmeyer tube. Magnetic stirrer heater (IKA C-MAG HS 7) is used to heat the solution. The solution was heated at 200 °C for 4 hours.

For the thiosulfate leaching tests, 6 gram of 200 mesh sample was used. a weighed amount of sodium thiosulfate pentahydrate (Merck) was dissolved in 10 mL Double-distilled water. Then the determined volume of ammonia solution (28% Merck) diluted with 10 mL of

Double-distilled water, and a weighed amount of copper (II) sulfate pentahydrate (Merck) was dissolved in 10 mL of Double-distilled water. The solution of all ingredients and sample were carried out in 100 mL Erlenmeyer tube. Magnetic stirrer heater (IKA C-MAG HS 7) is used to heat and stir the solution. The pH of the solution was then measured using pH meter and obtained a pH of about 10, according to the experimental reference (Mohammadi et al., 2017). Also, pulp density (S/L) ratio of 20% and stirring speed of 400 rpm were used in experiments.

The gold concentration in solutions was determined by inductively coupled plasma optical emission spectrometry (ICP-OES, Agilent Technologies 5100) with ICP Expert software version 7.3.1.9507. For gold ore characterizing were utilized using X-ray Flourescence (XRF, PANalaytical) and Laser-induced Breakdown Spectroscopy (LIBS) with SpectraGryph as analyzer software. Optical Microscope (Zeiss Primotech) was used to observe surface of ore.

Figure 1 shows presence of pyrite on a surface of ore, and The LIBS pattern of the ore is seen in Figure 2 also shows there is presence of a Pyrite (FeS). Both of it proves that the ore type is high sulfide ore. Presence of pyrite has a negative effect because it will reduce the effectiveness of thiosulfate leaching process because pyrite will make decomposition of thiosulfate faster (Feng & van Deventer, 2006).

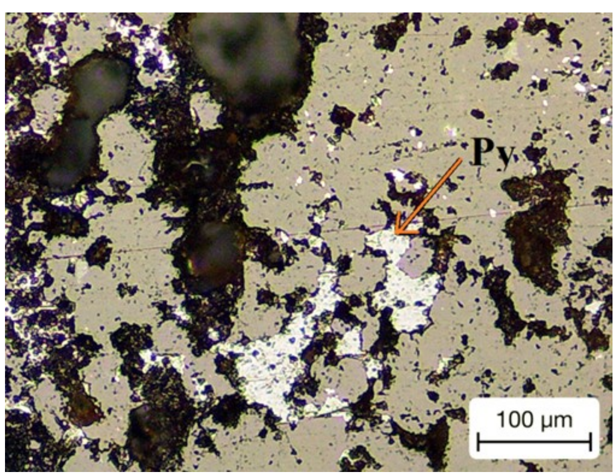

Figure 1. Result of Surface Ore Observation Using Optical Microscope

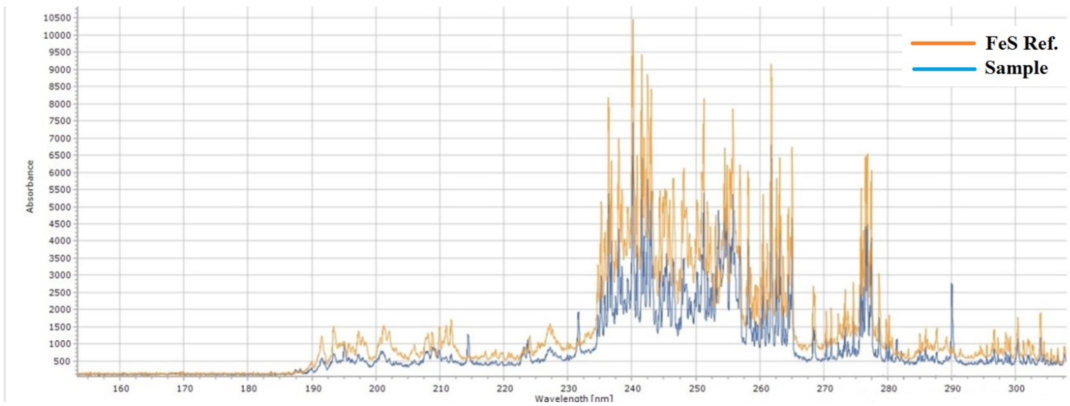

Figure 2. Laser-Induced Spectroscopy Result (Ore Spectrum vs. FeS Reference Spectrum)

Presence of pyrite can be reduced with pretreatment process, roasting is one of them. It will oxidized FeS become Fe_2O_3 (Marsden & House, 2006). Table 1 shows chemical analysis of roasted bolaang mongondow ore, it shows that percentage of sulfide become lower than Oxide that indicate roasting process successfully oxidized a pyrite. The gold content of the ore was determined as 0.27 ppm by aqua regia method.

Table 1. Chemical Analysis of Roasted Ore

Element	Si	Fe	S	K	Al	Ti	Ca	P	Na	Cl	Cu	Au
Conc. (%)	20.3	14.62	6.69	4.36	4.05	1.18	0.8	0.73	0.25	0.16	0.15	0.27 pm

RESULT AND DISCUSSION

1. Effect of Thiosulfate Concentration

The effect of thiosulfate concentration on gold extraction is shown in Fig 2. The gold extraction value was increased to about 62% using 0.1M thiosulfate after 2 h leaching. Increase of thiosulfate concentration has positive effect because thiosulfate is main reagent in this process that will make $Au(S_2O_3)_2^{3-}$ complex with Au (Breuer & Jeffrey, 2000). In higher thiosulfate concentration (0.2 M) there is slightly decline of gold percentage, this is caused by stability of $Cu(S_2O_3)_3^{5-}$ complex is higher than $Cu(NH_3)_4^{2+}$ with addition of thiosulfate concentration but same ammonia concentration (Navarro et al., 2002). It has negative effect because $Cu(NH_3)_4^{2+}$ is a catalyst in this reaction and when its existence decreased it will make reaction become slower.

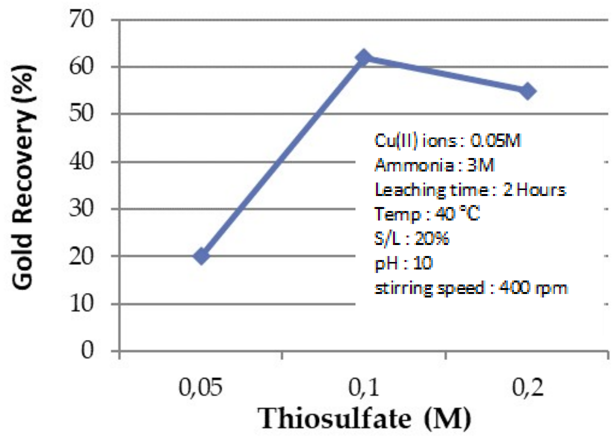

Figure 3. Effect of Thiosulfate on Gold Extraction

2. Effect of Temperature

Effect of temperature on gold extraction is shown in Fig. 3. The gold extraction increases when the temperature increases up to 40°C, while in higher temperatures the gold extraction value decreases. Temperature has a significant effect on the leaching process, namely on chemically controlled reactions. Similar findings were reported by other researchers regarding the effect of temperature (Altinkaya et al., 2020; Chen et al., 2022; Santoso et al., 2020). This conforms to the activation energy equation commonly known as the Arrhenius equation (Oraby, 2009).

$$k = Ae^{-E\alpha/RT} \tag{3}$$

Where:

k is the reaction rate constant,

R is the gas constant (8.31 J/K/mole),

T is the temperature, K,

A is the frequency factor

Eα is the activation energy.

It shows from Arrhenius equation that increase of temperature will increase reaction rate. In higher temperature (60°C), gold recovery lower than 40°C solution (62 % to 45%).

Higher temperature cause to increasing of ammonia volatility and loss. According to some research works (Gradov, 2013; Oraby, 2009) ammonia volatility and loss will affect stability of Cu (II) because Cu (II) cannot make $Cu(NH_3)_4^{2+}$ complex with ammonia and will make $Cu(S_2O_3)_3^{5-}$ instead. Loss of ammonia will also lead to passivation of thiosulfate on surface of ore because Au cannot make $Au(NH_3)^{2+}$ complex.

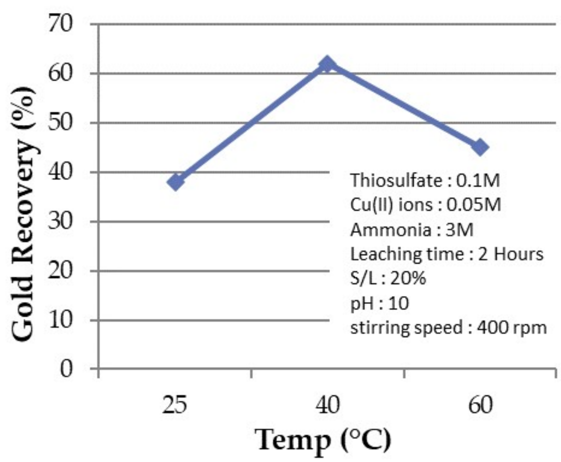

Figure 4. Effect of Temperature on Gold Extraction

CONCLUSION

According to the results of the present work, the following conclusions were obtained:

1. Gold extraction was increased with increase in thiosulfate concentration up to 0.1 M after 2 h leaching. Decrease in gold extraction was observed using 0.2 M thiosulfate concentration, it may cause by passivation of thiosulfate on surface of ore.
2. Gold extraction increased with increase in temperature up to 40°C. Excess increasing the temperature up to 60°C caused the gold extraction to decline because ammonia volatility and loss.

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