

## Article

# Optimization of Gold Dissolution Parameters in Acidified Thiourea Leaching Solution with Hydrogen Peroxide as an Oxidant: Implications of Roasting Pretreatment Technology

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**Abstract:** The nature of gold (Au) ore deposits plays an essential role in determining the best gold recovery method as an alternative to cyanidation, considering environmental concerns. Thiourea (Tu) leaching of gold is an alternative lixiviant for treating sulfide gold ores and concentrates. The present study investigated the leaching behavior of Au from sulfide gold ore concentrates obtained from the Cripple Creek site (Newmont operated mine) using acidified thiourea solution. The concentrates containing pyrite, K-feldspar, quartz, and gypsum as major minerals were roasted before leaching to break down complex sulfides and improve the gold's leachability. Parameters investigated include the leaching time (1–7 h), temperature (20–60 °C), pulp density (20–60 g/L), and thiourea concentration (20–60 g/L) to determine optimum conditions for Au dissolution. Hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) was used as an oxidizing agent and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) to maintain an acidic environment during the leaching process. The highest Au dissolution percentage of 96.2 was obtained after 6 h of leaching with 40 g/L thiourea at 60 °C, pH 1, and pulp density of 20 g/L. Based on the results presented in this work, the dissolution of gold with thiourea in an acidic solution would be a potential alternative to the traditional cyanide process, with the increase in understanding of the green leaching of Au from sulfide gold ores.

**Keywords:** thiourea leaching; concentrated sulfide ore; gold; hydrogen peroxide

## 1. Introduction

Gold (Au) is in high demand due to its wide range of applications such as electronic devices, aerospace, jewelry, and medicine [1,2]. Au offers high conductivity, ductility, and corrosion resistance, making it an ideal candidate for connections in electronic devices, and its ductility qualifies it to be used in coinage [3]. The average annual cost of gold in the United States was 1800 USD/oz in 2021, as reported in January 2022, which has slightly increased from 1261, 1272, 1395, 1774 USD/oz in 2017, 2018, 2019, and 2020, respectively [4]. The gold-ore deposits are scarce and are mainly associated with pyrite and other complex sulfide-bearing ores [5]. Hydrometallurgy is the best method to extract gold from the gold-bearing pyrite ore [6]. It is challenging to recover gold from its ore, especially from sulfide lower-grade ore. Therefore, the process should be optimized with respect to profitability, sustainability, and environmental benefits. The presence of refractory elements in the gold ores and their interaction with the gold particles make them difficult to process.

Refractory gold ores that contain sulfides and carbonaceous material need to undergo an oxidation pretreatment stage in order for the gold they contain to be susceptible to the subsequent leaching processes [7]. The oxidation roasting greatly influences refractory gold ore/concentrate as a common pretreatment process in gold extraction [8,9].

The most common leaching lixiviants used for gold recovery from sulfide gold ores are cyanide and mercury [10], but despite their low cost and efficiency in the gold mining industry, cyanide and mercury are toxic reagents harmful to human beings and the environment [11–14]. Halides, acidic thiocyanate, thiosulfate, and thiourea have been reported as potential substitutes for these toxic chemicals for leaching Au [15–21].

Studies have demonstrated that using thiourea to recover Au from ores is a promising alternative [22,23]. Various researchers have also utilized thiourea to recover Au from secondary resources such as the end life of electronic waste and flue dust [24–26]. Less than 85% of gold was leached when non-cyanide methods of extraction of gold from calcining samples were studied by some researchers [27–29]. Tu for leaching gold has many advantages, including high selectivity, faster gold-dissolution rate, easier handling, and low environmental pollution compared to conventional methods [16,30,31]. Tu gold-leaching is suitable for treating complex gold ores that are difficult to treat by cyanidation [32,33]. Acidic media of thiourea solution acidified with  $H_2SO_4$  could be considered the best in recovering gold, because it was proven to be non-corrosive and non-toxic compared to the currently used toxic chemical [34–36]. In acidic conditions, thiourea oxidizes Au to form a strong cationic complex called formamidine disulfide ( $Au[SC(NH_2)_2]_2^+$ ) (FDS) [37]. Thiourea is generally stable in an acidic solution with a pH of less than 4.5 [21,38]. Au leaching using Tu in alkaline conditions has been investigated and shown to be effective, but it still requires the presence of sodium sulfite ( $Na_2SO_3$ ) along with sodium persulfate ( $Na_2S_2O_8$ ) as the oxidizing agent. The presence of  $Na_2SO_3$  limits the stability of thiourea in alkaline media, which then affects the rate of Au dissolution [39]. Studies show that  $H_2SO_4$  is preferable to HCl for pH modification during the Tu gold-leaching, since it is easier to handle and relatively cheaper [40–42]. Some restricted conditions are required in thiourea leaching performance, such as control of redox potential, thiourea concentration, pH, oxidizing agent, and leaching time. However, these conditions are known to vary from ore to ore [30].

Oxidants could accelerate the leaching of gold from pretreated sulfide gold ore in thiourea solution.  $Fe_2(SO_4)_3$  is stated as the oxidant promoting the rapid dissolution of gold in thiourea [21]. Researchers have investigated the use of thiourea in the Au leaching process, but they mostly used Fe (III) as an oxidizing agent with a focus only on the mineralogical and physicochemical properties of the sample [26]. However, the use of  $Fe_2(SO_4)_3$  resulted in the formation of the iron sulfate–thiourea complex, and this led to excess Tu consumption [37]. Another disadvantage associated with using Fe (III) is that it leads to the accumulation of impurities due to co-dissolution, which makes the separation process difficult and results in the generation of secondary waste [43]. Although Fe (III) has been used as an oxidant in the leaching process, hydrogen peroxide ( $H_2O_2$ ) has been confirmed to be a more suitable alternative oxidant due to its ability to yield a high percentage of Au in solution [25]. Although thiourea leaching has been propounded in gold recovery from sulfide gold ore, the studies are still limited [44,45]. Nevertheless, these approaches required expensive equipment, high reaction temperatures, more chemical reagents, and low gold recovery efficiencies.

This study provides mineralogical information and conditions of leachability of Au from the concentrated sulfide of low-grade gold-ore deposit using Tu solution in the presence of  $H_2O_2$ . The study is mainly focused on the behavior of various leaching parameters, including thiourea concentration, leaching temperature and time, pH, and pulp density. All parameters were experimentally investigated for optimum conditions for maximum Au dissolution. The study validates the potential use of Tu to recover Au from sulfide gold ores with environmental benefits and cost incentives in the gold extractive industry.

## 2. Materials and Methods

### 2.1. Materials

The sample used in this study was a concentrate obtained after a flotation processing of low-grade gold-ore deposits from Newmont corporation, located in Colorado, USA. The chemicals used in the study were of analytical grade. Thiourea (>99.0%) and Hydrogen peroxide  $H_2O_2$  (30 wt.%) were purchased from Sigma-Aldrich, while  $H_2SO_4$  (95–98%) was purchased from J. T. Baker.

### 2.2. Methods

#### 2.2.1. Sample Characterization

The sample was pulverized to less than 45  $\mu m$  prior to mineralogical analysis. The particle analysis of concentrate samples was carried out by a Microtrac FlowSync (Microtrac Retsch GmbH, Haan/Duesseldorf, Germany). The investigation of mineralogical characteristics and phases present in the unroasted and roasted sample were identified using an x-ray diffractometer (XRD) (Malvern PANalytical, Westborough, MA, USA) with a  $Cu-K\alpha$  ( $\lambda = 0.1540$  nm) radiation source operated at 45 kV and 40 mA. The samples for XRD were prepared by putting sample powder on a sample holder, and the surface was well smoothed and flat and ready for the X-ray analyzer. The measurement of diffraction data was obtained at the scanning rate of 2 degrees/min with a 2-theta range of 5–90 degrees. The obtained diffractograms were interpreted using X'Pert HighScore Plus matched with the ICSD database (version 4.6a (4.6.1.23823)). Scanning electron microscopy in combination with energy-dispersive X-ray spectroscopy SEM-EDS (JEOL JSM-700F, Hollingsworth & Vose, East Walpole, MA, USA) was used to analyze the morphological and chemical composition of the sample. The samples were prepared by carefully dropping samples to the double-sided sticker fixed on the sample holder. To ensure that all excess particles were removed from the surface, air spray was used before the sample holder was mounted into the specimen. The sample was ready to be inserted into the SEM chamber for scanning [46]. Assay pulps were used for a suite of chemical analyses. These tests include gold content determined by Fire Assay with Atomic Absorption Spectroscopy (AAS) [47], C/S total for carbon and sulfur concentrations, and inductively coupled plasma mass spectrometry (ICP-MS, PerkinElmer, NEXION350x, Waltham, MA, USA) for elemental analysis. The samples were roasted at 500 °C for 2 h in a Muffle furnace (Mellen microthermal, Concord, NH, USA) under an oxygen atmosphere.

#### 2.2.2. Leaching Experiment

Leaching experiments were carried out in a closed borosilicate glass beaker of a 1000 mL with a magnetic stirrer on a hot plate. The sample size used in this study was less than 45 microns. For the redox potential measurement, Extech RE300 ExStik ORP Meter was used. The leach liquor was analyzed with ICP-MS to determine the chemical composition and the percentage of Au-dissolution. At the beginning of the experiment, a weighed quantity of ore and thiourea was charged into the beaker and an appropriate amount of deionized water was added. The pH was adjusted and fixed at 1 and monitored accordingly by dropwise addition of  $H_2SO_4$  to the mixture. A predetermined amount of hydrogen peroxide ( $H_2O_2$ ) was added to the ore/thiourea slurry. At predetermined time intervals during the leaching process, about 5 mL of the sample was taken out using a pipette and filtered on a vacuum pump with filter paper (<2.5  $\mu m$ ); the filtrate was diluted with 2% nitric acid depending on the concentration of the analyte and then subjected to the ICP-MS analysis. The Redox potential was measured after 30 min and at the end of each experiment. The effect of different leaching parameters such as leaching time (1–7 h), temperature (20–60 °C), pulp density (20–60 g/L), and thiourea concentration (20–60 g/L) was investigated. The random forest method was used to determine each parameter's significance according to its impact.

Each leaching experiment was repeated a minimum of 3 times and the average values were reported. For highly accurate experimental results, deionized (DI) water was used. The percentage of gold dissolved was calculated as follows:

$$Au(\%) = 100 (Au_{(solution)} / Au_{(solid)})$$

$$Au_{(solution)} = V_t * [Au]$$

$$Au_{(solid)} = Au \text{ content in the sample (mg/Kg)} * Feed \text{ (Kg)}$$

where  $Au_{(solution)}$  is the amount of gold in the thiourea solution,  $Au_{(solid)}$  is the amount of gold in the feed sample before the leaching process, and  $V_t$  is the total volume of solution (l), and  $[Au]$  is the amount of gold leached in solution in ppm. Figure 1 shows the experimental flowsheet followed in this study.

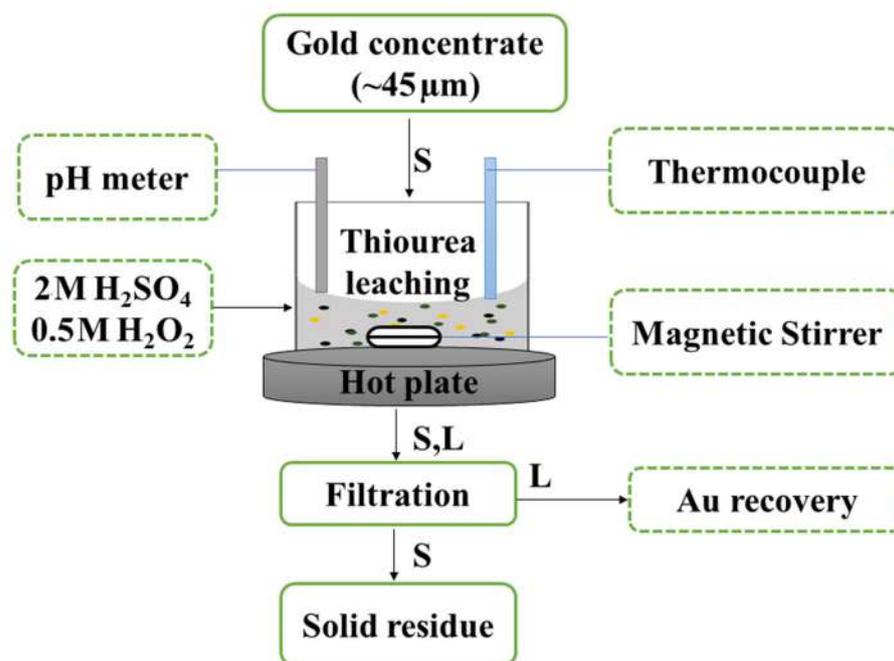


Figure 1. Flowsheet of Thiourea leaching process.

### 3. Results and Discussion

#### 3.1. Mineralogical Characterization of Materials

The material was first characterized in this study to understand its mineralogical and chemical composition. The particle size distributions show the size range in which the concentration of minerals with commercial value is most likely to occur. The Microtrac flowSync particle size analyzer was used to evaluate the ore sample's particle size distributions, and the results are represented as log-normal distributions in Figure 2. The particle size distribution of the gold concentrates results showed that the diameters D10, D50, and D90 values were 2.74, 10.59, and 104.0  $\mu\text{m}$ . The physical appearance of studied samples before roasting was in gray color, and after roasting at 500  $^{\circ}\text{C}$  for two hours became brown (Figure 3a,b). The results of the chemical characterization of the concentrated sample under investigation are presented in Table 1. From the table, the Au and Ag contents were determined as 17.04 ppm and <3.0 ppm, and the major component of the sample includes S (34.5%), Fe (29.26%), Al (3.49%), K (2.69%) and other minor components. Sulfur and iron content represent the presence of the  $\text{FeS}_2$  phase and confirmed in Figure 4a where the mineral phase analyzed for the sample before roasting shows the presence of pyrite ( $\text{FeS}_2$ ) as the major phase, along with K-feldspar, quartz, and gypsum. Figure 4b represents the roasted sample and sulfur converted into sulfur oxides. The phases identified were cristobalite ( $\text{SiO}_2$ ), hercynite ( $\text{FeAl}_2\text{O}_4$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), and ferric sulfate ( $\text{Fe}_2(\text{SO}_4)_3$ ).

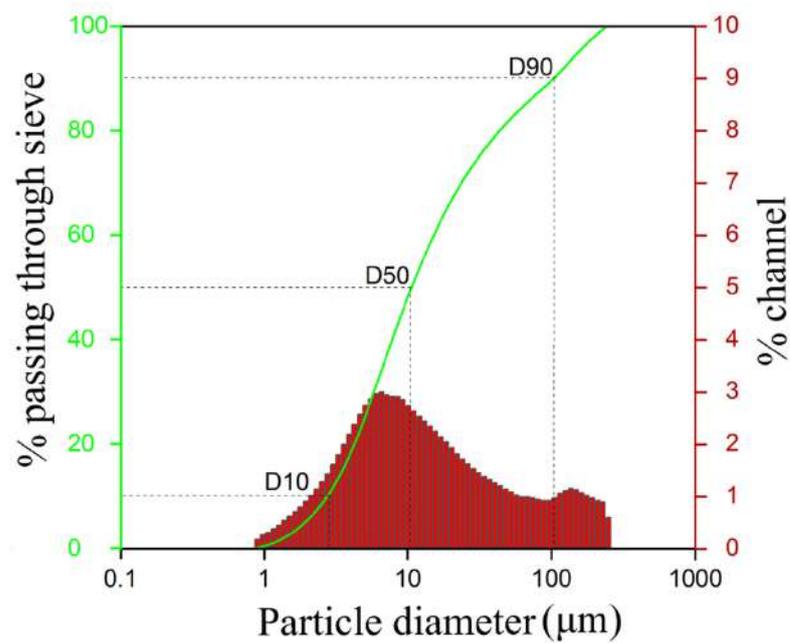


Figure 2. Particle size distribution of concentrated sulfide gold sample.

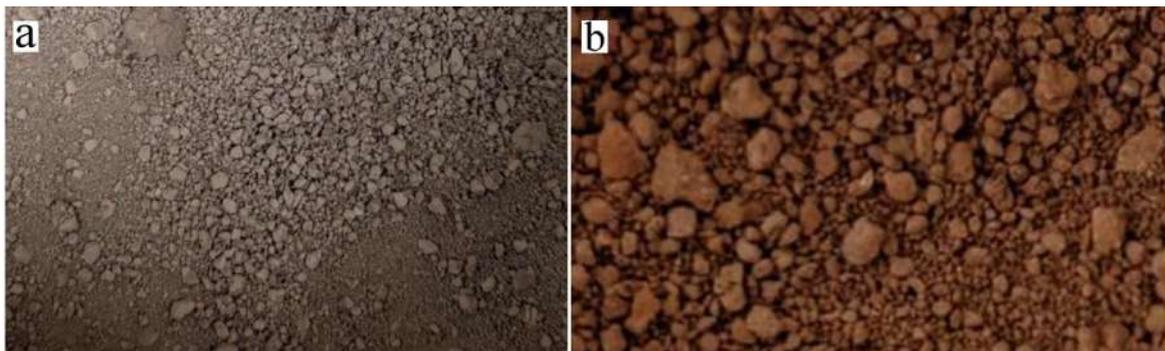


Figure 3. Image of unroasted sample (a) and roasted sample at 500 °C for 2 h (b).

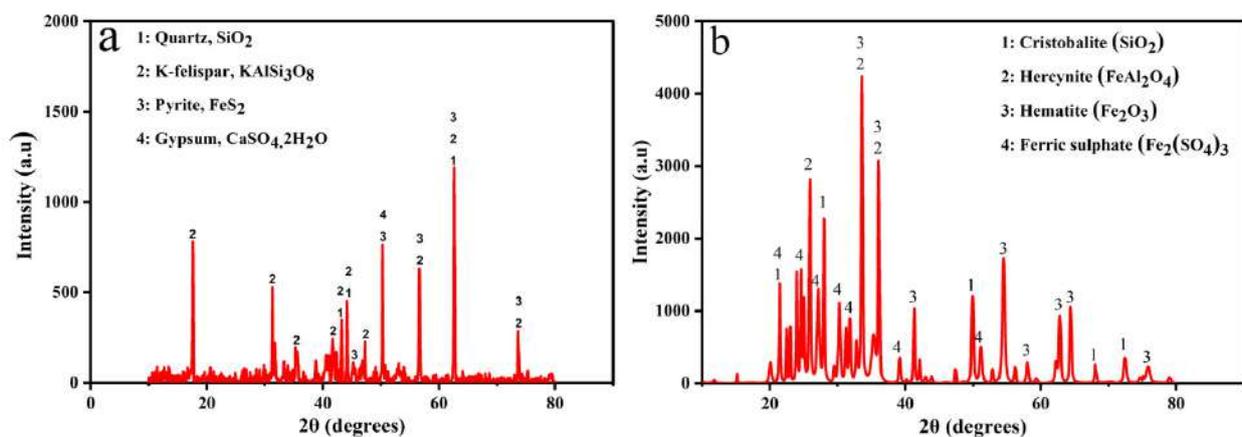
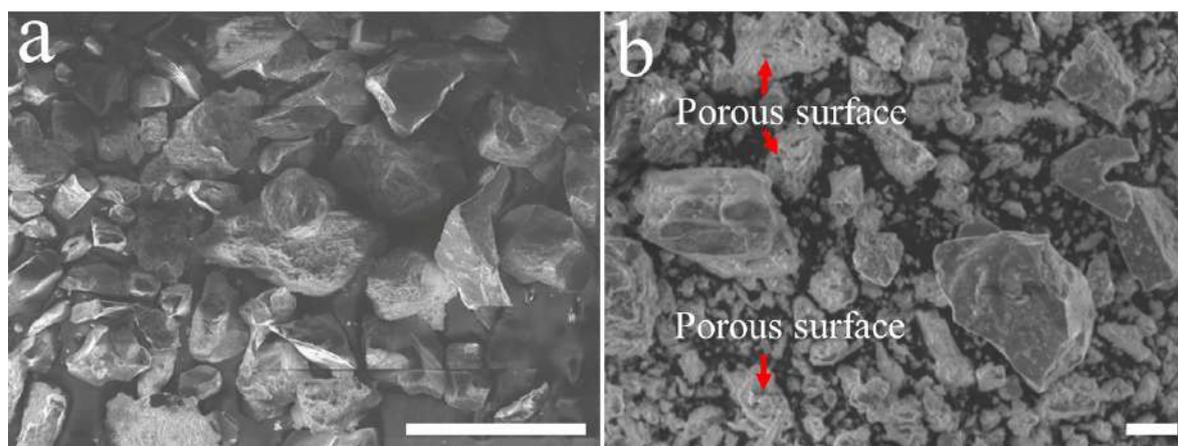
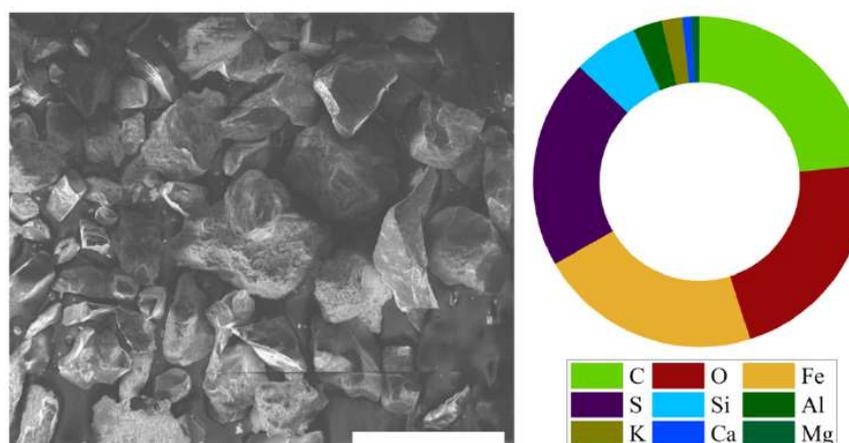


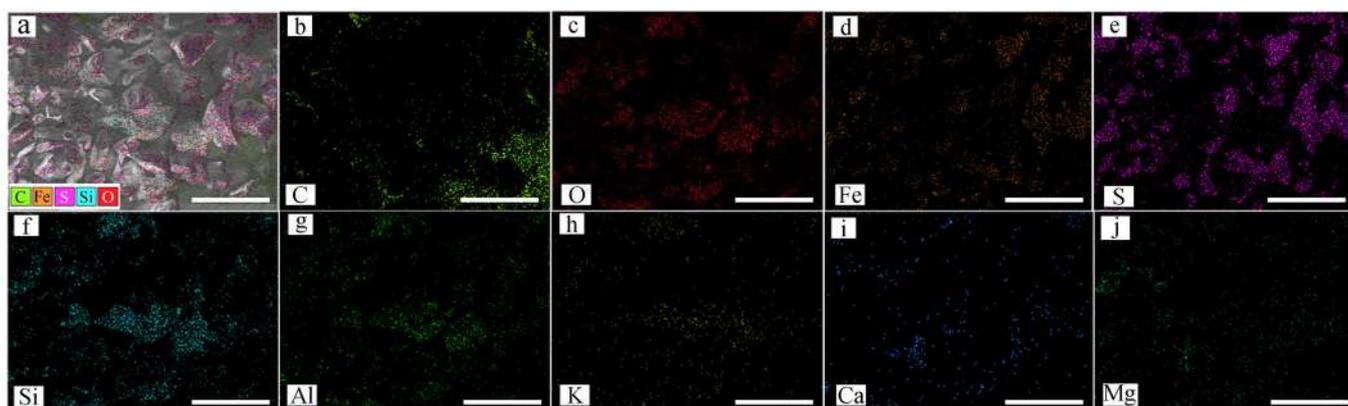
Figure 4. Crystallography of feed material, (a) as received sample, and (b) after roasting at 500 °C for 2 h.

**Table 1.** Chemical analysis of the concentrated and screened sulfide gold sample < 45  $\mu\text{m}$  (Au\*, Ag\*: ppm).

Element	Au*	C	S	Fe	Ag*	Al	As	Ba	Be	Ca	Co	Cr
Content (wt.%)	17.04	0.75	34.51	29.26	<3.00	3.49	0.05	0.06	<2	1.19	0.01	0.01
Element	K	Mg	Mn	Zn	Mo	Na	Ni	Pb	Sb	Sr	Ti	Tl
Content (wt.%)	2.69	0.75	0.12	0.08	0.06	0.59	0.008	0.03	0.003	0.09	0.43	0.14

The SEM micrograph in Figure 5a (before roasting) indicates that the sample particles are smooth while Figure 5b (after roasting) shows the rough surface and all have irregular shapes. We also carried out surface and element analysis of the concentrated sulfide gold sample using SEM-EDS, and the results are presented in Figure 6. The semi-quantitative analysis from EDS results represented by the pie chart in Figure 6 showed that the components of the studied sample were C, O, Fe, S, Si, Al, K, Ca and Mg with 23.4%, 21.7%, 21.6%, 20.5%, 6.3%, 2.8%, 2.0, 0.9 and 0.8%, respectively. The major sample components were Fe (21.6%), S (20.5%) and Si (6.3%) in Figure 6 and clearly mapped as depicted in Figure 7a. As the amount of sulfur and iron content was significantly high in the sample, this implied that gold was hosted by sulfide in the form of pyrite (Table 1, Figures 6 and 7a). This was confirmed by the XRD analysis in Figure 4a, which shows the presence of K-feldspar, pyrite, quartz, and gypsum phases with smaller peak intensities.

**Figure 5.** SEM micrograph, (a) before roasting, and (b) after roasting at 500  $^{\circ}\text{C}$  for 2 h.**Figure 6.** SEM-EDS of sulfide concentrated gold sample; the scale bar is 250  $\mu\text{m}$ .



**Figure 7.** EDS element maps of sulfide concentrated gold sample: (a) combined image, (b–j) images of individual elements, the scale bar is 250  $\mu\text{m}$ .

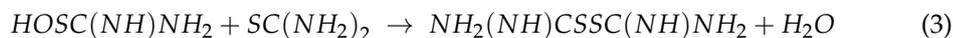
Knowledge of the nature of the sample is a quintessential step in determining the best processing route for any mineral ore [48]. The mineralogical information of the concentrated sulfide sample was conducted using various methods (AAS, XRD, and SEM), since gold particles with the associated impurity properties information provide the image of processing routes. Hence, the optimization depends on physical–chemical knowledge of the gold-hosted rock behavior. The information from the quantitative analysis provided the nature of the studied sample [49]. The type of gold hosted by sulfide matrices like pyrite and arsenopyrite is known as refractory ores [50]. Previous studies showed that the direct leaching of refractory ore leads to low gold extraction, and that pretreatments are required to unlock sulfide encapsulation [7]. Therefore, the sample was roasted for 2 h at 500  $^{\circ}\text{C}$  and the phases were identified as quartz in the form of cristobalite ( $\text{SiO}_2$ ), hercynite ( $\text{FeAl}_2\text{O}_4$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), and ferric sulfate ( $\text{Fe}_2(\text{SO}_4)_3$ ), as depicted in Figure 4b. This facilitates the increase in gold extraction due to the rise of the pore (Figure 5b) and desulfurization process than unlock the gold from its host.

In the roasting process of sulfidic gold concentrate, the major components such as pyrite and/or arsenopyrites easily convert into iron oxides, sulfate, sulfur oxides, arsenic oxides, and arsenate when heated at 500  $^{\circ}\text{C}$  [45]. Figure 4b, hercynite ( $\text{FeAl}_2\text{O}_4$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), and ferric sulfate ( $\text{Fe}_2(\text{SO}_4)_3$ ) formed after roasting the sulfide-concentrated gold sample. During the roasting process under controlled conditions, S and As components could oxidize to  $\text{SO}_2$  and  $\text{As}_2\text{O}_3$ . This change contributes to unlocking the fine gold from sulfidic gold ore and concentrates by enhancing the leachability of gold in the hydrometallurgical process.

### 3.2. Leaching Gold from Roasted Concentrated Sulfide Gold Sample

The Au leaching with Tu solution works very well in acidic conditions and the parameters tested are given in Table 2. It has been revealed that the pH range experimentally from 1 to 2 results in optimum gold dissolution [37,51]. Furthermore, there is more metal liberation for particle sizes less than 75 microns, increasing the contact surface and leading to the fast reaction between thiourea and gold particles, and high Au dissolution [25]. The present study used the roasted concentrated sulfide sample with particle size below 45 microns for leaching, this served to increase the surface area for enhanced Au dissolution. Optimization of the leaching parameters may minimize the consumption of the lixiviant and the cost. This work conducted a series of Au leaching experiments to establish the best conditions for high Au dissolution. When thiourea is added to the acidic solution, gold dissolves in the form of  $[\text{Au}(\text{CS}(\text{NH}_2)_2)_2]^+$  as shown in Equation (1) [26,30]. The chemistry of gold thiourea-leaching with  $\text{H}_2\text{O}_2$  as an oxidant to accelerate the reaction is shown in Equa-

tion (2) [52], and this oxidant oxidizes thiourea and forms FDS  $[NH_2(NH)CSSC(NH)NH_2]$  as displayed in Equation (3) [53]. The FDS formed facilitates Au dissolution from the solution.



**Table 2.** Parameters tested in the acidified thiourea leaching tests, leaching conditions, Au recoveries at pH: 1.

Time (h)	Temperature (°C)			Tu Concentration (g/L)				Pulp Density (g/L)			Tu/H <sub>2</sub> O <sub>2</sub>				Agitation Rate %			
	Values	25	40	60	20	30	40	60	20	40	60	0.4	0.6	0.8	1.2	300	400	500
				<b>Average</b>														
1	55.2	69.1	84.2	51.5	61	74.5	60.5	84.2	68.3	64.4								
2	61.3	72.5	80.4	57.2	60.4	80.2	67	80.4	72.5	68.1								
3	70.3	76.1	87.3	60.1	69.9	93.9	73.2	87.3	73.3	69.1								
4	75.9	80	93.2	66.5	76.9	91.6	78	93.2	71.5	71								
5	78	80.8	94.9	74	80.7	87.9	79.8	91.4	71.6	72.1								
6	79.9	82.9	91.9	96.2	86.1	95.6	94.4	90.9	79.9	75.2	73	82	87	92	81	95	96	
7	81.9	83.9	93.7	96.1	86.5	94.8	93.6	92.2	86.9	82.7								
				<b>Standard deviation</b>														
1	7.8	6.6	5.8	1.8	0.6	8.3	2	7.3	3.1	3.1								
2	7.5	3.3	5.3	4.2	0.6	0.8	4.2	3.9	3.3	1								
3	3	6.1	4.8	2.8	0.9	3.5	7.3	9	4.4	2.7								
4	7.4	5.1	6.7	4.2	4.8	4	3.7	5.2	5.5	6.9								
5	6.2	2.8	7	1.4	0	3.3	4.6	4.9	2.3	3.3								
6	2.2	3.5	6.8	3.1	5	3.8	3.2	6.8	8.8	4.6								
7	3.7	5.6	0.9	1.4	3.2	1	4.7	1.2	4.4	3								
	<b>Potential at the beginning and at the end of experiment hours (mv)</b>																	
0.5	253	234	221	255	231	244	239	247	250	249								
6	228	200	200	108	209	203	220	230	216	239								

The effect of agitation speeds of 300, 400 and 500 rpm were tested. In 6 h at 60 °C, Au dissolution was determined as 80.6%, 95.3%, and 95.8% for 300, 400, and 500 rpm, respectively. It was found that when the agitation speed was 400 and 500 rpm, there was not much difference in gold dissolution, whereas at low stirring speed the dissolution drops to 80%. Therefore, the Au recovery increases with the increase in the agitation rate.

### 3.2.1. Effect of Thiourea Concentration

The influence of different concentrations of thiourea in acidic media on the leaching of gold was investigated, and the results are presented in Figure 8a. The Tu concentration varied from 20–60 g/L while the other experimental conditions were fixed at 60 °C, 0.5 M H<sub>2</sub>O<sub>2</sub>, pH 1, and the pulp density was 20 g/L. At Tu concentration of 20 g/L, it was observed that the gold dissolution rate was low (52%) after 1 h of leaching and increased gradually with leaching time. After 5 h of leaching there was a sharp increase in the dissolution rate from 75% to a maximum value of 96.2% at a leaching time of 6 h. The gold dissolution rate increased with thiourea concentration in all experiments and reached high peak values after 6 h; however, the thiourea concentration of 30 g/L had a lower peak dissolution rate compared to the rest. For Tu concentration of 40 g/L, a higher dissolution rate could be achieved even at a lower leaching time of 3 h.

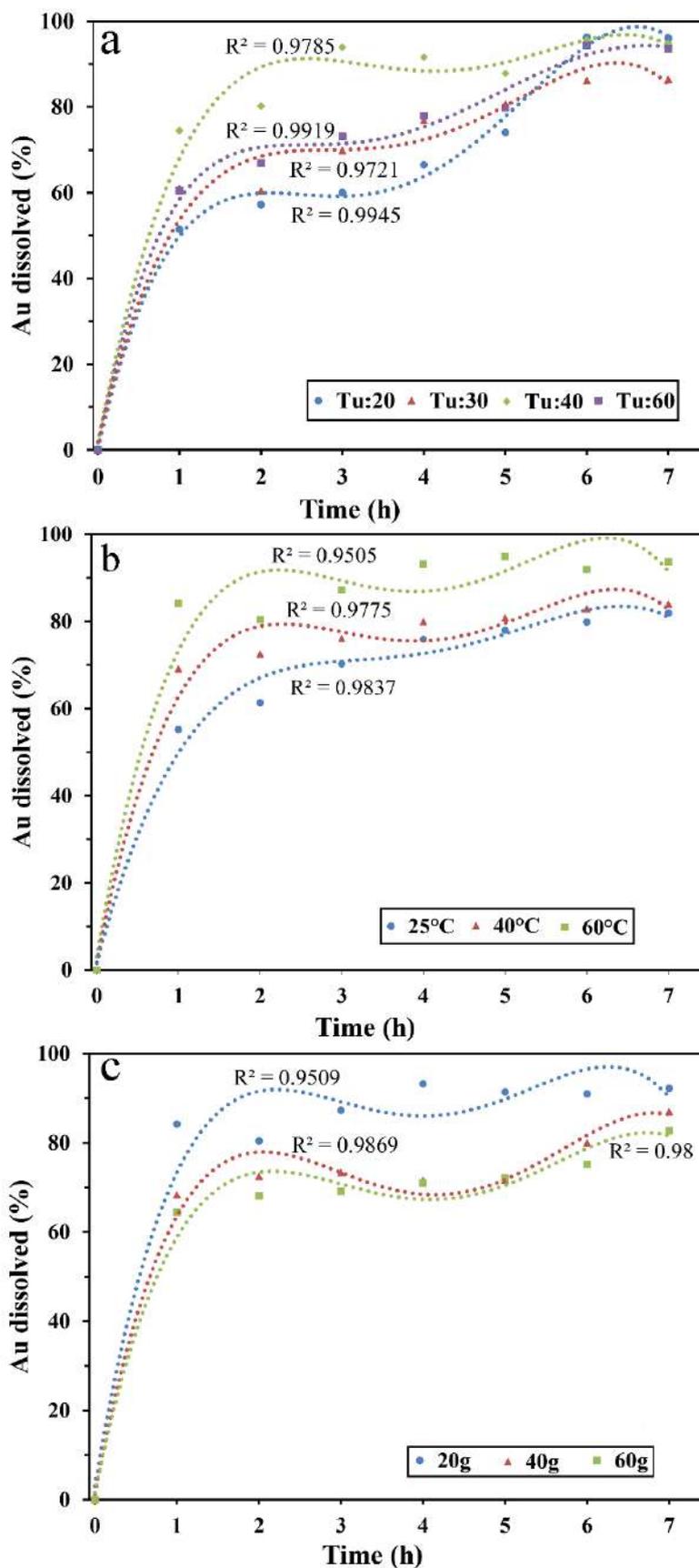


Figure 8. The effect of (a) thiourea concentration, (b) temperature and (c) pulp density on Au dissolution. (Experiment condition 0.5 M H<sub>2</sub>O<sub>2</sub>, pH: 1).

At this concentration, over 70% Au dissolution was realized from a leaching time of 1 h and beyond, while for the other concentrations the same dissolution rate was achieved after a leaching time of 4 h. Although the highest Au dissolution percentage could be seen after 6 h for a Tu concentration of 20 g/L, on average the Au dissolution was higher at 40 g/L. Therefore, the Tu concentration of 40 g/L was taken as the optimum concentration and used for other experiments. In the previous studies where Tu/H<sub>2</sub>O<sub>2</sub> was used in gold extraction, the recovery rate was varied from 69 to 98% from the secondary resources, and the highest obtained from primary resources was 91.94% at 60 °C [25,54]. This study's results show a recovery rate of 96.2%, which is better and in the same range as the recovery rates shown above.

### 3.2.2. Effect of Temperature

The pulp temperature is an important factor in gold dissolution due to its effect on the solubility of the soluble elements in the solution and the dissolution time. Figure 8b shows the effect of temperature on Au dissolution for leaching time from 1–6 h, where we observed an increase in Au dissolution with the time for the different temperature ranges considered. The experimental condition was carried out using 40 g/L thiourea, 0.5 M H<sub>2</sub>O<sub>2</sub>, 2 M H<sub>2</sub>O<sub>4</sub>, and the pulp density was 20 g/L. The highest gold dissolution rate was achieved at a temperature of 60 °C in 5 h of leaching, but after this it decreased. At the lower temperature, the reactions were slow and required more time to promote the high Au dissolution rate. However, above 60 °C the studies showed that high evaporation might occur during chemical processing reactions [55]. The increase in dissolution rate at the beginning was observed and then dropped down at 2 h. This is due to the increase in the temperature after adding H<sub>2</sub>O<sub>2</sub>, as H<sub>2</sub>O<sub>2</sub> decomposes into oxygen (exothermic decomposition) and waste.

The dissolution rate at 40 °C and 60 °C was faster than that at 25 °C. According to the obtained results, Au dissolution increases with further time and temperature increases. The high evaporation may cause a drop in pH up to 0.6 or less, leading to a conspicuous decrease in the dissolution rate [20]. The temperature was able to dissolve 95% of the gold at the optimum time of 5 h Figure 8b, and a decrease followed this in Au dissolution percentage. The temperature in this study did not exceed 60 °C because hydrogen peroxide easily decomposes above 60 °C [56].

### 3.2.3. Effect of Pulp Density

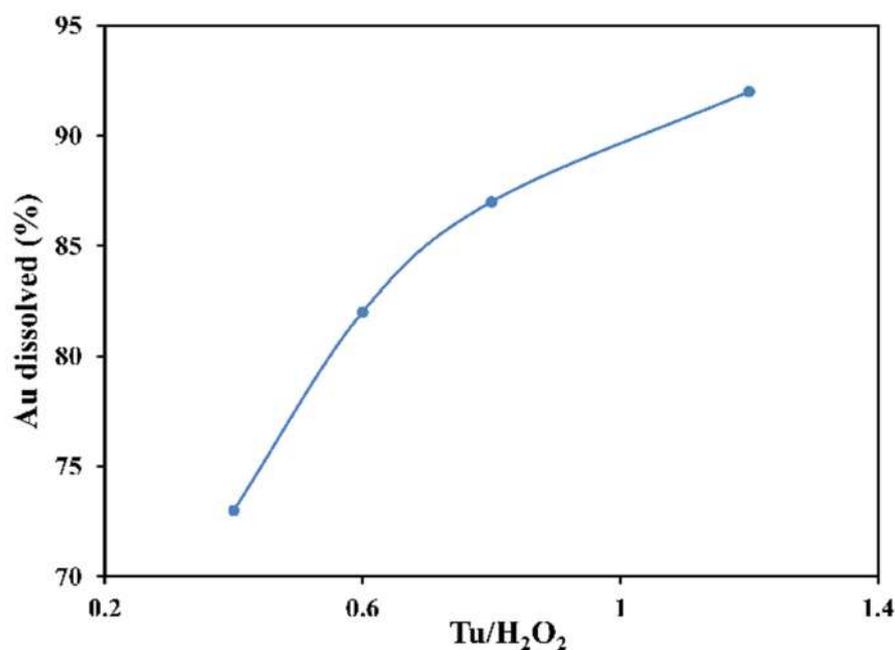
Pulp density is another important parameter that can influence the Au leaching process. The influence of the pulp density on Au dissolution rate was studied by varying the amount of the sample in Tu solution such that the pulp density varied from 20 g/L to 60 g/L. The experiment was conducted at a temperature of 60 °C, 0.5 M H<sub>2</sub>O<sub>2</sub>, pH 1, and the thiourea concentration of 40 g/L. Figure 8c shows the effect of the pulp density on the Au dissolution rate. The figure clearly indicates that the highest Au dissolution rate was achieved at a pulp density of 20 g/L after 4 h of leaching. The Au dissolution rate decreased when the leaching time exceeded 4 h. For the pulp densities of 40 g/L and 60 g/L, the Au dissolution rate increased with the leaching time, and the dissolution rate followed a similar trend. In general, there was a decrease in the dissolution rate with the pulp density, which implies that the dissolution rate decreased with an increase in Au solid content in the leaching tank. The leaching efficiency of gold increases with a lower solid/liquid ratio; as the pulp density increases, there is a need for greater thiourea concentrations [44]. Therefore, the pulp density of 20 g/L was selected as the best value to be used for subsequent experimental sets.

The mass balance was investigated with the experimental condition of the pulp density of 20 g of the sample at 60 °C, 0.5 M H<sub>2</sub>O<sub>2</sub>, pH 1, 40 g/L of thiourea at 6 h. After filtration, the solid residue was dried at 100 °C in the oven. The leached amount may also contain other chemical elements that can be totally dissolved (Ag) or partially dissolved (for

example Fe, Ca, Al) in thiourea solution [55]. In this study Ag content was very low, less than 3 ppm Table 1. The Au dissolution at this condition was 95%.

#### 3.2.4. Effect of Tu/H<sub>2</sub>O<sub>2</sub> Ratio

Figure 9 depicts the effect of oxidant-thiourea ratio on gold dissolution during leaching at (pH: 1, 6 h, pulp density: 20 g/L). The figure shows that the increase in the Tu/H<sub>2</sub>O<sub>2</sub> ratio augmented the amount of gold in the solution. However, from the literature, the excess oxidant concentration results in high thiourea consumption and thiourea decomposition, thus limiting the dissolution of gold and making the process expensive [57]. In this experiment, the concentration of hydrogen peroxide was kept constant with the variation in Tu concentration. The higher Au percentage obtained was 92.2% at 1.2 Tu/H<sub>2</sub>O<sub>2</sub> (Figure 9), which is a better condition than the preceding studies showing their optimum ratio at 2 [51].



**Figure 9.** The effect of Tu/H<sub>2</sub>O<sub>2</sub> upon Au dissolution (experimental condition: pH: 1, 6 h, pulp density: 20 g/L).

#### 3.2.5. Effect of Agitation

To determine the effect of agitation speeds, 300, 400, and 500 rpm were tested. Gold dissolution in 6 h at 60 °C was recorded as 80.6%, 95.3%, and 95.8% for 300, 400 and 500 rpm, respectively. Figure 10 shows that when the agitation speed was 400 and 500 rpm there was little difference in gold dissolution, whereas at low stirring speed the dissolution dropped to 80%. Therefore, the Au recovery increased with the increase in the agitation rate.

#### 3.2.6. Effect of Potential on Tu Leaching

The thiourea-gold leaching complex is more stable at an electrochemical potential in the range between 0.2–0.6 V at low pH [43]. Gold could leach to significant dissolution in thiourea solutions if the potential is well maintained. An oxidant such as H<sub>2</sub>O<sub>2</sub> or Fe<sup>3+</sup> is required to maintain the potential to the appropriate range. In this investigation, the initial measured potential (240 mV) decreased with the increase of time (Table 2). At the end of the experiment, the highest recorded potential measured was 108 mV. The average potential noted varied between 140 mV and 255 mV. In this case, the high Au dissolution was obtained at a low potential. This indicates that in the reaction the thermodynamic driving forces are powerful [58].

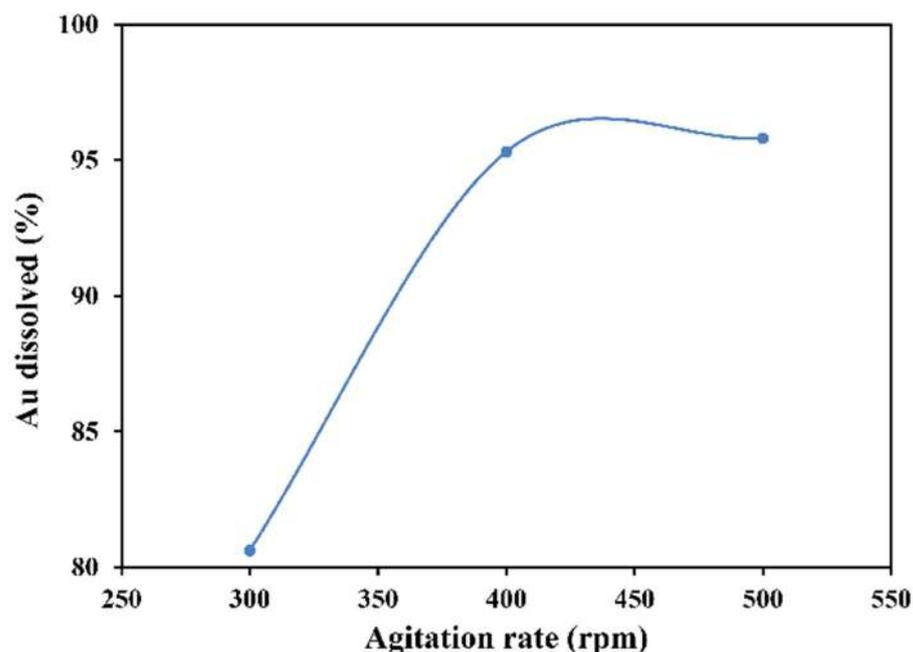


Figure 10. Diagram of Au dissolved % vs. agitation rate rpm.

#### 4. Conclusions

This study provides mineralogical information and leachability of Au from a sulfide gold concentrate calcine (17.04 ppm) of low-grade Au ore using Tu solution in the presence of  $H_2O_2$ . The major phases of the unroasted concentrate were pyrite, K-feldspar, quartz, and gypsum; the roasted sample major phases were cristobalite, hercynite, hematite, and ferric sulfate. Roasting increases the specific surface area and pore diameter of minerals, which is advantageous to accelerating the kinetic process of gold leaching and improving gold leaching efficiency confirmed by the transition of the phases. The result obtained, after varying several leaching parameters, was 96.2% of Au dissolution obtained at the optimum condition of 40 g/L of thiourea, pH ~1, temperature of 60 °C, with the relevant Tu/ $H_2O_2$  ratio of 1.2 at 6 h and 400 rpm. The variation in gold extraction rate was associated not only with the mineralogical and physicochemical properties of the sample, but also with the ability to control necessary conditions.

Based on the experimental results with the aid of the random forest method, the significance of different factors in the dissolution of Au is as follows: Time > Thiourea concentration > Solid/liquid ratio > Temperature, according to their impact. The results obtained from this work revealed that using Tu and hydrogen peroxide as an oxidant is viable for gold recovery and is more suitable than using cyanide leaching with environmental and economic incentives. Thus, the thiourea consumption rate and extraction of gold from pregnant solutions should be investigated in future studies to extract gold from sulfide gold ore. Apart from excelling on floatation concentrates and secondary resources, this method can be developed and applied to lower-grade ores.

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