Extraction of Gold From Boulangerite Ore by Ammonium Thiocyanate (NH₄SCN)

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Abstract. In this study extraction of gold from Boulangerite ore (Pb,Sb,S₁₃) via hydrometallurgical leaching process was studied. Boulangerite ore sample was collected from Shishy Valley, Chitral, Khyber Pakhtunkhwa, Pakistan. The influence of various parameters on the extraction of gold from boulangerite ore was investigated i.e., the amount of ammonium thiocyanate, leaching time, particle size of ore and temperature. Ore with particle size in the range 100-300 mesh gave satisfactory yield values. An increase in amount of ammonium thiocyanate and boulangerite ore, leaching time and particle size resulted in an increase in the amount of leached gold. At optimal conditions the extraction of gold was observed to increase from 32% to 80.69%.

Keywords: boulangerite ore, gold leaching, ammonium thiocyanate

Introduction
The demand for gold is constantly increasing due to its widespread use in electronic devices, jewellery and as economy. Consequently, its recovery from the relevant ores has become more important over the last few decades. Solvent extraction of gold from leach liquors has been studied extensively using ion pair or solvent extractants e.g., dibutylcarbitol (diethylene glycoldibutyl ether), 4-methyl-2-pentanone (methyl isobutyl ketone; MIBK) and di (2-ethylhexyl) phosphoric acid (Akita et al., 1996). Solvent extraction has been used for the recovery of metals by means of hydrometalurgical process but this method is not used commercially for the purification and concentration of gold (Caravaca et al., 1996). Gold can be absorbed and eluted effectively using either strong or weak base anion exchange resins. Loading strong base resins with gold cyanide is an ion exchange mechanism involving the exchange of resin anions. Weak base resins typically have about one-half of the loading capacity than the strong base resins, but it needs an increase in the pH of the solution (Hariss et al., 1992). Using a mixture of amines and neutral organophosphorus derivatives has also been reported e.g., gold (I) has been extracted using primene 81R in cyanide media (Caravaca, 1994).

Cyanidation is an important process for extraction of gold from its ore. Gold cyanide complexes are formed as a result of gold dissolution in aerated cyanide solution. A large proportion of gold can be recovered from sulphide ores that are soluble in cyanide solution (Jeffrey and Breuer, 2000). However, a series of environmental accidents at various gold mines around the world has received widespread concern over the use of cyanide as a leaching reagent. In majority of these cases, cyanide from processing operations entered the environment either by leakage through tears and/or punctures in protective heap leach liners, or by spillage from over flowing solution ponds or tailings storage areas (Hilson and Monhemius, 2006). For this reason, gold leaching by cyanidation has been banned in many regions of the world in recent years. Also a cyanidation process usually takes more than 24 h. Therefore, a slow gold leaching is often a problem of cyanidation process. In summary, high toxicity of cyanide, slow leaching kinetics and low gold extraction from refractory ores constitute the main problems of cyanide leaching (Orgul and Atalay, 2002).

Thiourea leaching is the most suitable alternative for cyanidation. The main difference between them is the use of different extracting agents. A large proportion of anionic thiourea is converted into formamidine disulphide (FDS) with the help of an oxidizing agent, which reacts with gold in an excessive thiourea medium. In order to minimize thiourea consumption, the solution pH and potential values (mV) must be controlled (Gonen, 2003). Another non-cyanide leaching reagent thioulsulphate has received much attention in recent years. Ammonium thioulsulphate is an inexpensive nontoxic reagent. Acceptable leaching rates have been achieved using thioulsulphate in the presence of ammonia with cupric ion acting as the oxidant. For example a total of

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1. 24 million tonnes low-grade refractory gold ore has been processed with ammonium thiosulphate at Newmont’s operation near Carlin, Nevada, USA (Wan and Levier, 2003).

It was found that thiocyanate was an environment friendly reagent for gold leaching and more stable than thiourea in acidic solution. It was preferred to be used under weakly acidic conditions with a pH of 1.0 to 2.0 (Kholmogorov et al., 2002). Additionally, ammonium thiocyanate was cheaper than thiourea, sodium cyanide in commerce. However, gold leaching with thiocyanate was slower than with cyanide (Li, 2012). Based on these previous studies, a low toxicity and low-corrosiveness solution of ammonium thiocyanate was thus used in the current work as lixiviant to extract gold from boulangerite ores.

Materials and Methods

Boulangerite ($\text{Pb}_3\text{Sb}_2\text{S}_11$) ore was collected from Shishy Valley, Chitral (35.84°N:71.78°E), Khyber Pakhtunkhwa, Pakistan. Semi-quantitative EDS detected the presence of 52.43 wt% Pb, 24.85 wt% Sb, 19.76 wt% S, 1.51 wt% Cu and traces of Au (15 ppm) in as-mined boulangerite ore sieved through a 200 mesh at Materials Research Laboratory (MRL), University of Peshawar.

The leaching experiments were carried out in a fume hood (LFH-120 SCI, LabTech). 3.0 g of weighed dried sample was placed in a 500 mL glass beaker. A deionized water of 20 mL with a pH of 1.5 adjusted using 2.5 M $\text{H}_2\text{SO}_4$ was then put in the flask. After that 2.0 g of thiourea were added to the flask, unless specified otherwise. The mixture solution was stirred at temperature of 60 °C at a speed of 300 rpm by an electromagnetic stirrer with a Teflon coated stirring bar and a LED indicator showing the stirring speed. Aqueous samples were prepared in order to investigate the effect of various parameters i.e., leaching time, particle mesh size of the ore, amount of ammonium thiocyanate and amount of boulangerite ore. All the samples were analyzed for gold content using anatomic absorption spectrometer (AAS 700, Perkin Elmer, USA) in Centralized Resource Laboratory (CRL), University of Peshawar, Pakistan.

The percentage of gold extraction was calculated according to the following equation.

$$\text{Au extraction (\%)} = \frac{[\text{Au}] \times V}{W_{\text{Au}}} \times 10^2$$

where:

- $W_{\text{Au}}$ is the weight of gold in milligram in the as-mined$_{\text{Au}}$ sample:
- $[\text{Au}]$ is the concentration of gold from the filtrate in mg/L;
- $V$ is the volume of filtrate in liter.

Results and Discussion

Effect of leaching time on the extraction of gold.

Figure 1 shows the observed variation in the amount of gold at 200 mesh particle size as a function of leaching time.

The experimental results demonstrated that the extraction of gold increased from 33 to 36% by increasing leaching time from 1 - 3 h; therefore, in the present study, maximum gold extraction was observed for 3 h leaching duration. Table 1 shows various parameters set for gold extraction in the present study.

The observed increase in the amount of extracted gold with time may be due to the relatively more mass loss (gangue) of the sample.

The effect of particle size of boulangerite ore on Au extraction.

Figure 2 shows the observed variation in amount of extracted Au as a function of particle size of the boulangerite ore. It was observed that the amount of Au extracted increased from 32 to 38% with a decrease in the particle size of the ore from 100 mesh (149 μm) to 300 mesh (44 μm).

![Graph showing variation of gold extraction with time](image)

**Fig. 1.** Variation in the amount of extracted gold$_{\text{Au}}$ from boulangerite ore as a function of leaching time (Experimental parameters: 60 °C, 300 rpm, ammonium thiocyanate 2g, boulangerite ore 3g, distilled water 20 mL).
The observed increase in the amount of extracted Au may be due to the diffusion of particles which allows relatively more thiourea to interact with Au particles leading to more leaching and hence, an increase in Au concentration.

**Effect of ammonium thiocyanate amount.** Figure 3 shows the observed variation in the amount of extracted Au from boulangerite ore as a function of ammonium thiocyanate content. Au extraction was observed to increase from 32% to 51% as a result of increasing ammonium thiocyanate amount from 3-6 g. The observed increase may be due to preferential adsorption of the ammonium thiocyanate onto the gold with increase in thiocyanate content.

**Effect of boulangerite ore (\( \text{Pb}_3\text{SB}_5\text{S}_{11} \)) amount.** Figure 4 shows the effect of ore amount on the extraction of gold from boulangerite ore and it dispatch that the amount of extracted Au increased from 32- 40% with an increase in the amount of ore from 3- 7g.

**Table 1.** Quantitative data regarding the extraction of Au under the optimum conditions established in the present study

<table>
<thead>
<tr>
<th>Boulangerite ore (g)</th>
<th>Ammonium thiocyanate (g)</th>
<th>Distill water (mL)</th>
<th>Temp (°C)</th>
<th>RPM</th>
<th>Particle size (mesh)</th>
<th>Time (h)</th>
<th>Filtrate amount (mL)</th>
<th>Au extraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>7</td>
<td>6</td>
<td>30</td>
<td>60</td>
<td>300</td>
<td>300</td>
<td>3</td>
<td>25</td>
<td>80.69</td>
</tr>
</tbody>
</table>
**Bulk production of the concentrate.** The results of the experiment conducted under the optimum conditions established in the present study (i.e. 60 °C leaching temperature, 3 h leaching time, 300 rpm, 300 mesh, 6 g ammonium thiocyanate, 7g boulangerite ore) for leaching of Au are summarized in Table 1. These parameters enabled the extraction of 80.69 % of Au from boulangerite ore.

**Conclusion**

This study demonstrated the suitability of the ammonium thiocyanate process for the gold recovery from boulangerite ore (Chitral) at laboratory scale. The effect of various factors on the extraction of Au from examined boulangerite ore containing 52.43 wt% Pb, 24.85 wt% Sb, 19.76 wt% S, 1.51 wt% Cu and 1.45 wt% traces of Au (15 ppm) was investigated. The present experimental results demonstrated that the extraction of Au mainly depended on the concentration of free ammonium thiocyanate available for leaching i.e. higher Au extraction was achieved by increasing the amount of ammonium thiocyanate. Similarly the extraction of Au also increased with an increase in leaching time, particle size of the ore and the amount of boulangerite ore used. Employing the optimum experimental conditions established in the present study, the amount of extracted Au was 80.69 %. Thus ammonium thiocyanate can be commercially used for Au extraction from boulangerite ore.

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