



Dual lixiviant leaching process for extraction and recovery of gold from ores at room temperature



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ABSTRACT

A comprehensive investigation of Au leaching from gold ores with a solution containing thiourea and ammonium thiocyanate was performed. A gold ore A with 18 g/t of Au and 1.15 wt.% of sulfur and another gold ore B with 55.7 g/t of Au and 11.67 wt.% of sulfur were used. The percentages of extracted Au were 57%, 66% and 95% for the leaching solutions containing thiourea only, ammonium thiocyanate only and ammonium thiocyanate plus thiourea, respectively, under optimal conditions. There were some interactions among thiourea, ammonium thiocyanate and ferric ion, which affected gold extraction greatly. The addition of thiocyanate reduced greatly the consumption of thiourea and the process cost accordingly. The optimal concentrations of thiourea, ammonium thiocyanate and ferric sulfate for ore A leaching were 0.13 M, 0.78 M and 0.028 M, respectively. The optimal pH, temperature, time, ratio of liquid to solid, agitation speed and particle size for ore A leaching were 1.5, 21 °C, 6 h, 10, 250 rpm and 54–74 μm, respectively. Au extraction for ore A was 95% under above optimal conditions. Au extraction was 97% when sulfur content of ore B was 1.48 wt.%. The preferential consumption of ferric ions by pyrite over by gold was the main reason for resulting in the decrease of gold extraction. Desulfurization is necessary to achieve a high gold extraction for this leaching system of thiourea-thiocyanate-Fe³⁺. After Au extraction, about 90% of Au was recovered as insoluble metallic Au by cementation using 5 g/L of Al powder. The cementation filtrate was reused to leach Au ore and about 91% of Au was extracted after pH adjustment. Thus, thiocyanate and thiourea could be reused, which reduced the process cost. The leaching speed of Au of this method was more than 4 times faster than that of cyanidation process. This process concerning extraction and recovery of Au from Au ore was effective, fast, environmentally friendly and also likely economical.

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1. Introduction

Cyanide leaching has been the predominant process for gold extraction from mineral resources for more than a century (Hilson and Monhemius, 2006; Li et al., 2012b; Örgül and Atalay, 2002). The major reasons for the acceptance of cyanide over the other lixivants include higher chemical stability, lower cost and better understood chemistry (Ofori-Sarpong and Osseo-Asare, 2013). Another reason is that the adsorption of gold cyanide complex onto the most widely used adsorbent, activated carbon, and the elution of gold from activated carbon have been intensively studied (Ofori-Sarpong and Osseo-Asare, 2013). 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 the 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 overflowing 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 (Grosse et al., 2003). In addition, problems can often occur when leaching gold from complex ores such as those containing sulfur, arsenic, copper, lead, zinc, manganese, telluride and carbonaceous ores (Grosse et al., 2003; Örgül and Atalay, 2002), resulting in poor recoveries of gold. A cyanidation process usually takes more than 24 h (Li and Miller, 2006, 2007; Örgül and Atalay, 2002). Thus, a slow gold leaching is often a problem of cyanidation process. In summary, high toxicity of cyanide, slow leaching kinetics and low gold extraction for refractory ores constitute the main problems of cyanide leaching.

For these reasons, considerable efforts have been made to find alternative gold lixivants (Hilson and Monhemius, 2006; Li et al., 2012b; Ofori-Sarpong and Osseo-Asare, 2013). A variety of low-toxic and low-corrosive gold extraction processes have been studied for this purpose (Chandra and Jeffrey, 2005; Feng and Van Deventer, 2007; Hilson and Monhemius, 2006; Jeffrey and Brunt, 2007). Thiosulfate was found to undergo disproportionation or oxidation in aqueous solutions, forming various other sulfur species including sulfite, sulfate, di-, tri- and higher polythionate during the leaching process, thus resulted in an excessive consumption and accordingly it was found that it was very difficult to

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find an operable regime (Li et al., 2012b; Senanayake, 2007). Thiourea as a new type of non-cyanide reagent has received extensive attentions (Li and Miller, 2007; Poisot Díaz et al., 2012). It was used under acidic conditions using Fe^{3+} as an oxidizing agent (Tremblay et al., 1996; Whitehead et al., 2009). The use of thiourea resulted in a faster leaching of gold compared to cyanide leaching (Örgül and Atalay, 2002). However, thiourea was not stable in acidic solution and often decomposed in the presence of oxidizing agent (Li and Miller, 2006; Li et al., 2012b,d). Toxicity data on thiourea indicates that it has a high threshold limit for mammals and a lethal dose of 10 g/kg for humans (Mensah-Biney, 1997). Thiourea has been used in the treatment of thyroid diseases in human for years and is currently considered non-carcinogenic to humans (Mensah-Biney, 1997). Thiourea is actually classified in class/group 3 (The agent is not classifiable as to its carcinogenicity to humans) of the International Agency for Research on Cancer (IARC) classification for carcinogenicity. This is an authoritative and independent source, as the IARC is part of the World Health Organization (WHO). Therefore, thiourea can be considered much safer than cyanide. It was found that thiocyanate was an environmentally friendly reagent for gold leaching (Kholmogorov et al., 2002; Kuzugüdenli and Kantar, 1999). It was preferred to be used under weakly acidic conditions with a pH of 1.0 to 2.0 (Kholmogorov et al., 2002). Thiocyanate is more stable than thiourea in acidic solution (Barbosa-Filho and Monhemius, 1994; Broadhurst and Du Perez, 1993; Li et al., 2012a). Additionally, ammonium thiocyanate was cheaper than thiourea in commerce. However, gold leaching with thiocyanate was slower than with cyanide (Li et al., 2012c). Based on the above previous studies, a low-toxicity and low-corrosiveness mixture solution of thiourea and ammonium thiocyanate (Hilson and Monhemius, 2006) was thus used in the current work as lixiviants to extract gold from gold ores, considering a combination of the unique advantages of both. In addition, a recycling use of thiourea and thiocyanate was also studied. Although both thiourea and thiocyanate have been studied to leach gold from pure gold disk (Yang et al., 2011), their use as lixiviants of a real gold ore has never been reported before to the best of our knowledge. This work performed using two types of real gold ores was quite different from those using pure gold disks as samples (Li and Miller, 2007; Li et al., 2012b,c; Örgül and Atalay, 2002; Poisot Díaz et al., 2012; Safarzadeh et al., 2012; Yang et al., 2010, 2011), because an enhanced interaction between thiourea and ferric ion catalyzed by some compounds contained in real ores has been reported (Li and Miller, 2006).

2. Experimental

2.1. Materials

A gold ore A with 18 g/t of Au and 1.15 wt.% of sulfur and another gold ore B with 55.7 g/t of Au and 11.67 wt.% of sulfur were used in this work. They were procured from the region of Inner Mongolia of China. The gold ores A and B were crushed, dried, ground and sieved. Some samples with various particle sizes were thus obtained and used in this work. The gold ore A with particle size fraction of 49–74 μm was more frequently used in this work unless otherwise specified. The sample was dried at 105 °C for 2 h and kept in a desiccator. The chemical compositions of ores A and B were listed in Table 1. The solid reagents of thiourea, ammonium thiocyanate and ferric sulfate of analytical grade were used in this work. The Al powder with particle size of 200 ± 20 mesh (about 74 μm) and analytical grade was used in this work.

Table 1
Main chemical compositions of gold ores A and B^a.

Compositions	SiO ₂	Fe	Al	K	Ca	S	Mg	Au
Content (wt.%) (gold ore A)	63.21	13.46	4.78	2.76	1.17	1.15	1.04	18 (g/t)
Content (wt.%) (gold ore B)	46.30	23.97	0.40	0.73	0.16	11.67	0.12	55.7 (g/t)

^a Contents were obtained for gold ores A and B with particle size ranging from 48–74 μm .

2.2. Procedures

2.2.1. Desulfurization of gold ore B

10.0000 g of weighed sample of gold ore B with a particle size of 49–74 μm was placed in a quartz boat. The boat was put inside a quartz tube. The boat was located at the center of the tube furnace (Fig. 1). An argon gas was passed inside the tube at a flow rate of 200 mL/min for 15 min. Then the furnace temperature was raised to 600 °C in an atmosphere of argon gas. After that, an oxygen gas replacing argon gas was passed inside the tube at a flow rate of 200 mL/min for a preset time ranging from 0.25 h to 2.5 h. After that the furnace was naturally cooled to room temperature. In this way, the treated samples of gold ore B with different sulfur contents were obtained. The Eschka analytical procedure described in Chinese state standard method (GB/T214 (Chinese State Standard), 2007) was used to determine total sulfur in the samples. Only 0.2000 g of the treated sample was used for determination of sulfur content, the rest of the sample was used for gold leaching.

2.2.2. Gold leaching procedure

10.0000 g of weighed dried sample was placed in an Erlenmeyer flask. A deionized water of 100 mL with a pH of 1.5 adjusted using 2.5 M H₂SO₄ was then put in the flask. After that 1.0 g of thiourea, 6.0 g of thiosulfate and 1.12 g of ferric sulfate were usually added to the flask, unless specified otherwise. The mixture solution was stirred at room temperature of 21 °C at a speed of 250 rpm by a electromagnetic stirrer with a Teflon coated stirring bar and a LED indicator showing the stirring speed. After agitation for 15 min, the sample solution was measured for ORP (oxidation–reduction potential) with an ORP electrode. The agitation of the solution was stopped temporarily when the solution ORP was measured. The ORP value at this time was referred to as initial ORP. After agitated for 6 h, the sample solution was measured for ORP again. The solution ORP at this time was called as final ORP. After that the sample solution was filtered through Whatman GF-A membrane. The filtrate was measured for Au concentration. The solid residue was dried at 105 °C and then weighed for measuring the weight loss of sample after leaching.

For the leaching experiment at the temperature other than room temperature, the Erlenmeyer flask was immersed in a bath filled with a circulating refrigerant R134a provided by a refrigerated and heating circulator (F12-ED, JULABO, Germany).

The percentage of extracted gold was calculated based on Eq. (1).

$$\text{Au extraction(\%)} = \frac{[\text{Au}] \times V}{W_{\text{Au}}} \times 10^2 \quad (1)$$

where W_{Au} was the weight of gold in milligram from the sample; $[\text{Au}]$ the concentration of gold from the filtrate in mg/L; V the volume of the filtrate in liter.

The mass loss of sample after leaching was calculated based on Eq. (2).

$$\text{Mass loss(\%)} = \frac{W_0 - W_R}{W_0} \times 10^2 \quad (2)$$

where W_0 and W_R were the weights of dried ores before and after leaching, respectively.

A redox electrode, using Ag, AgCl/KCl (222 mV/S.H.E.) as a reference, was used to measure the solution ORP. All ORP values are given relative

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