Minerals Engineering 24 (2011) 115–121

Contents lists available at ScienceDirect

Minerals Engineering

journal homepage: www.elsevier.com/locate/mineng

Thiosulphate leaching of gold in the presence of carboxymethyl cellulose (CMC)

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ARTICLE INFO

Article history:
Received 17 August 2010
Accepted 15 October 2010
Available online 18 November 2010

Keywords:
Gold ores
Sulphide ores
Leaching
Surface modification

ABSTRACT

Carboxymethyl cellulose (CMC) was introduced as a surfactant in the leaching of pure gold as well as a sulphide ore using ammoniacal thiosulphate medium. Competing with thiosulphate anions to complex with the cupric ion at the axial coordinate sites, CMC in small quantities reduced the consumption of thiosulphate in the ammoniacal thiosulphate leaching system. The dissolution of pure gold in thiosulphate solution was enhanced in the presence of CMC, and this beneficial effect became more pronounced with an increase in the CMC concentration up to a certain level. Extraction rates of gold and silver from the sulphide ore were improved greatly, while the consumption of thiosulphate was reduced in the presence of CMC especially at a higher CMC concentration. The enhanced leaching performance was attributed to the stabilisation of thiosulphate, likely reduced leaching passivation and the dispersion of mineral particles in the slurry system. Addition of CMC facilitates satisfactory gold extraction at low reagent consumption in thiosulphate leaching.

1. Introduction

Leaching of gold with ammoniacal thiosulphate solutions has been studied extensively as an alternative technology to the traditional cyanidation process (Kerley, 1981, 1983; Zipperian et al., 1988; Abbruzzese et al., 1995; Marchbank et al., 1996; Wan, 1997; Wan and Brierley, 1997; Thomas et al., 1998; Aylmore and Muir, 2001; Muir and Aylmore, 2004). Acceptable gold leaching rates can be achieved in the presence of ammonia with Cu^{2+} acting as the oxidant. Gold leaching in ammoniacal thiosulphate solutions is an electrochemical reaction, with the constituent half reactions being the oxidation of gold to gold thiosulphate, and the reduction of the cupric tetra-amine to the cuprous thiosulphate, as expressed in Eqs. (1) and (2), respectively (Breuer and Jeffrey, 2000):

$$Au + 2S_2O_3^{2-} \rightarrow Au(S_2O_3)_{2^{3-}} + e^- \quad (1)$$

$$Cu(NH_3)_4^{2+} + 3S_2O_3^{2-} + e^- \rightarrow Cu(S_2O_3)_3^{5-} + 4NH_3 \quad (2)$$

One of the major problems in thiosulphate leaching is the high consumption of thiosulphate. This is mainly caused by thiosulphate decomposition in solutions containing copper, as shown in the following equation:

$$2Cu(NH_3)_4^{2+} + 8S_2O_3^{2-} \rightarrow 2Cu(S_2O_3)_3^{5-} + 8NH_3 + S_4O_6^{2-} \quad (3)$$

In general, the thiosulphate leaching conditions reported in the literature are severe with high reagent consumption. Zipperian et al. (1988) reported a loss of up to 50% of thiosulphate in ammoniacal thiosulphate solutions containing copper. Early work at higher concentrations of reagents reported that the formation of copper sulphide and sulphur layers on gold particles would hinder their dissolution. Bagdasaryan et al. (1983) and Pedraza et al. (1988) observed a sulphur layer as well as copper sulphide in a thiosulphate–copper system. Electrochemical impedence spectral studies also showed that gold passivation could occur in the absence of copper (MacDonald, 1990).

From an economic point of view, it is required that the leaching should be carried out under low reagent concentrations over extended periods where reagent consumption is low. In order to achieve this, three key problems need to be solved, i.e. stabilising thiosulphate, maintaining copper ions at low concentrations in solutions to catalyse gold oxidation with low ammonia concentrations, and reducing passivation to allow gold leaching over extended periods. The ore mineralogy affected thiosulphate leaching of gold and thiosulphate consumption, and sulphide minerals were observed to retard the gold leaching and to catalyse the degradation of thiosulphate (Feng and Van Deventer, 2001; Van Zijll De Jong, 2005). Efforts were made to minimise thiosulphate consumption by reducing the concentration of thiosulphate in solution (Cao et al., 1992). Nevertheless, dilute solutions of thiosulphate (<0.01 M) were found to decompose more rapidly than concentrated solutions (>0.1 M) (Dhawale, 1993). Alternatively, sulphite was added to the leach solution to stabilise thiosulphate (Kerley, 1981, 1983). However, sulphite significantly retarded the thiosulphate leaching of gold by reducing Cu^{2+} in solution. More

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doi:10.1016/j.mineng.2010.10.007

recently, additives were used to enhance the thiosulphate leaching of gold via the stabilisation of thiosulphate (Xia et al., 2003; Alonso-Gomez and Lapidus, 2009; Feng and Van Deventer, 2010a,b). Despite the importance, little has been done to prevent the formation of passivation layers at gold surfaces and stabilise copper ions at low concentrations in solutions in the presence of sulphides. It is expected that the modification of the surface properties of particles with surfactants may affect the adsorption of copper and sulphur species on the particles, and hence the leaching passivation and the stability of copper in solutions.

Carboxymethyl cellulose (CMC), as a natural polymer, has been used extensively in the minerals industry to depress hydrophobic gangue minerals by rendering their surfaces hydrophilic in flotation (Bakinov et al., 1964; Shortridge et al., 2000; Morris et al., 2002). CMC is also a polyelectrolyte reagent, which renders mineral particles highly charged. Therefore, the addition of CMC could prevent the aggregation of fine particles, which is beneficial to the leaching system. In addition, the modification of gold surfaces with CMC may hinder the formation of sulphur species at the surfaces. Furthermore, CMC could reduce the decomposition of thiosulphate through the interactions between the carboxymethyl and hydroxyl groups and Cu^{2+} ions. It is the purpose of this study to investigate to what extent CMC affects thiosulphate leaching of pure gold as well as a sulphide ore at low reagent concentration through modifying the properties of gold and mineral particle surfaces.

2. Experimental work

2.1. Materials

Gold foils (99.99% Au, thickness 0.2 mm) were used in the pure gold leaching tests with a surface area of about 20 mm^2. The gold foils were polished with 0.1 µm monocrystalline diamond paste (Electron Microscopy Sciences), washed with acetone twice, rinsed with distilled water and swept with lint-free paper. A new gold foil was used for each leaching experiment. It should be noted that gold in general exists in the form of Au/Ag alloys with about 5% Ag in gold ores. According to previous experience, the leaching behaviour of gold followed a similar trend for pure gold and Au/Ag alloys. CMC (sodium salt, viscosity of 1500–2500 cP for 1% aqueous solution at 20 °C, BDH Laboratory Supplier) was prepared freshly with the addition of distilled water. All other chemicals were analytical or reagent grade.

Pure pyrite, quartz and haematite samples were obtained from Geological Specimen Supplies, Australia. A sulphide gold ore was obtained from a Newcrest gold mine with a particle size of 80% passing 75 µm. A rotary splitter was used to obtain representative samples of the ore for experimental use. Quantitative XRD was used to determine the mineralogy of the sulphide ore. An elemental analysis was performed by digestion of the ore. The mineralogical and elemental analyses are shown in Table 1, which lists only the elements considered most important in the study.

Table 1
Chemical and metallurgical analysis of the sulphide ore.

Quantitative XRD analysis		Elemental analysis	
Mineral	Content (mass%)	Element	Content
Albite	23.8	Cu (%)	0.07
Arsenopyrite	0.1	Fe (%)	5.27
Calcite	1.1	Total S (%)	3.9
Chalcopyrite	0.2	Sulphide S (%)	3.0
Dolomite	13.8	Au (mg/kg)	4.3
Muscovite	6.6	Ag (mg/kg)	2.0
Pyrite	5.4	Ni (mg/kg)	64
Quartz	48.1	Zn (mg/kg)	23

A diagnostic leaching test using thiosulphate was conducted previously on a similar sulphide ore (Feng and Van Deventer, 2010c) where it was shown that the gold is mainly "free" and complete recovery can be achieved with thiosulphate at severe reagent conditions. In the current study lower reagent concentrations will be used. Similarly, cyanidation tests showed that almost 100% gold recovery was possible at high cyanide concentrations, so that this sulphide ore is not refractory.

2.2. Analytical techniques

Elemental concentrations in solutions were determined by ICP-OES, involving the oxidation of sulphur species to stable sulphates prior to the analysis. After oxidation by hydrogen peroxide, solutions were acidified by aqua regia and boiled to ensure complete conversion of the metal species to the chloride form. For silver analysis, the oxidised solutions were only acidified by HNO$_3$. The thiosulphate concentration was determined by iodometric method with the addition of acetic acid (10% solution) for eliminating the interference of the cupric tetra-amine complex with the titration. A platinum electrode (M21Pt, Radiometer) was used to measure the mixed solution potential with a double-junction reference electrode (Ag/AgCl, saturated KCl, Orion) to avoid the interference of thiosulphate with the reference electrode. All potentials were given with respect to the Standard Hydrogen Electrode (SHE). Zeta potential was measured by the acoustic method in a Colloidal Dynamics (Model Acoustosizer II) instrument. Samples were ground to a particle size of -20 µm and sonicated for 1 min with an ultrasonic probe (UP 200S, dr. Hielscher GMBH) for surface cleaning. Ten gram of cleaned samples was added to 200 mL 0.01 M NaCl electrolyte solutions prepared with deionised water. Readings started from about pH 12 to 2 with the addition of NaOH or HCl. The point of zero charge (PZC) could shift due to the formation of particle aggregates after the initial several measurements.

2.3. Leaching tests

Experiments for pure gold leaching were performed in a 250 mL reactor with a sampling port using a magnetic stirrer. A leach solution of 200 mL was used with desired concentrations of CMC. The gold foils were suspended in the upper part of the leaching reactor with a nylon thread, ensuring no contact with the reactor wall during leaching. The stirring speed was maintained at 400 min^{-1}. The extent of gold dissolution was calculated based on the dissolved gold mass per m^2 of the gold foil surface.

The leaching of the sulphide ore was performed in a 1.5 L baffled PVC reactor using an overhead flat-bladed impeller. The reactors were open to the air through the sampling ports. A leach solution of 1.0 L was added to the sulphide ore of 400 g. A natural pH of about 10.3 was maintained in the experiments, due to the buffering effect of NH_4^+/NH_3. Leaching tests were carried out at a rotation speed of 250 min^{-1} and a temperature of 25 °C in a water bath. Samples were taken continuously at certain intervals during a total retention time of 48 h. The samples were immediately subjected to the subsequent iodine titration and oxidation for ICP analysis. The extent of extraction of gold or silver was the percentage of leached gold or silver from the sulphide ore. Duplicate experiments were conducted with only average results being reported, due to the standard deviations of all the tests being within 3%.