



# Effect of minerals on the stability of gold in copper ammoniacal thiosulfate solutions – The role of copper, silver and polythionates



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## ABSTRACT

The effect of some typical oxide and sulfide minerals, as well as activated carbon, on synthetic gold thiosulfate leach solutions was examined. The presences of pyrite and goethite in a synthetic ore slurry have a significant effect on the adsorption and stability of gold, silver, copper and polythionates in ammoniacal thiosulfate solutions. Adsorption of tetrathionate in particular leads to significant gold and silver losses. The adsorption of copper on mineral surfaces inhibited gold and silver losses, even in the presence of tetrathionate. However, goethite was found to strongly adsorb copper(II). It is concluded that gold extraction from natural ores is lower than that obtained with cyanide, partly as a result of sorption or precipitation reactions. Furthermore, the presence of silver in solution catalyses the precipitation of a mixed copper/silver/gold sulfide in ammoniacal thiosulfate solutions containing tetrathionate.

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## 1. Introduction

With increasing regulatory scrutiny of new gold projects and a lowering of what are considered acceptable levels of cyanide discharges, world industry attention has focused on ammoniacal thiosulfate as a relatively cheap and non-toxic reagent alternative to cyanide for leaching gold ores. The establishment of an economic and environmentally acceptable option for extracting gold without the use of cyanide would seem highly desirable, and in some cases, essential.

Previous research has shown that by maintaining sufficient concentrations of thiosulfate, ammonia, copper and oxygen in the leach solution under suitable  $E_h$  and pH conditions gold can be readily extracted with modest reagent consumption (Aylmore and Muir, 2001a, 2001b; Li et al., 1996; Molleman and Dreisinger, 2002; Muir and Aylmore, 2002; Wan, 1997). Despite considerable research into the optimum conditions for gold dissolution by thiosulfate, the maximum gold extractions generally obtained remain some 5–25% less than those achieved using cyanide.

In addition, the levels of reagent concentrations for effective leaching rates in ores are normally at least 50 times greater than that used in the cyanide leaching system. Furthermore, extending leaching times in an attempt to increase gold extraction can result in the loss of gold from solution, due to the formation of increased concentrations

of thiosulfate degradation products such as tetrathionate, sulfide and trithionate. This ultimately reduces the effectiveness of thiosulfate as an alternative lixiviant to cyanide for leaching gold. There have been extensive studies investigating additives to enhance gold and silver leaching, some beneficial and others detrimental (Alonso-Gomez and Lapidus, 2008, 2009; Feng and van Deventer, 2010a, 2010b, 2010c, 2011a, 2011b; Jeffrey et al., 2008; Xia and Yen, 2005, 2008). These additives either affect the residual copper(II) concentration and  $E_h$ , modify surface reactions with gold by catalytic action and/or prevent the formation of or remove passivating layers on gold surfaces (Senanayake, 2013).

Whilst thiosulfate leaching is currently attractive for carbonaceous ores, a number of issues and problems must be resolved before a general process can be demonstrated as practical and economic for typical oxidised gold ores (Muir and Aylmore, 2005).

The main reasons for lower gold extraction in thiosulfate compared with cyanide systems undoubtedly include precipitation or adsorption of species from solution associated with particular minerals in the ore (Muir and Aylmore, 2002). This paper describes a systematic study of the effects of selected oxide and sulfide minerals as well as activated carbon on the thiosulfate leaching system. In particular, gold adsorption and stability, the effect of copper, silver and polythionates are examined. This work was carried out under the auspices of the industry funded AMIRA P420 Gold Processing Technology project.

Although carbon has a low affinity for  $\text{Au}(\text{S}_2\text{O}_3)_2^{3-}$  (Gallagher, 1989) activated carbon has a high specific surface area relative to other minerals and could still reduce gold recoveries by sorption. Activated carbon has been identified as having similar properties to that of 'preg-robs' gold from cyanide solutions in carbonaceous ores (Ibrado and

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Fuerstenau, 1992). Evidence of the influence of some minerals on gold and silver dissolution rate has been presented in the literature particularly for pyrite and haematite (e.g. Feng and van Deventer, 2002; Zhang and Jeffrey, 2008). However, the study on the stability of gold and silver in ammoniacal thiosulfate solutions in the presence of minerals is limited.

## 2. Experimental

Particular oxide and sulfide minerals were chosen as representative of those commonly present in gold ores and mixed with finely ground quartz to produce a synthetic ore. The minerals investigated, together with the values for their specific surface areas and the mixtures with quartz used are indicated in Table 1. The compositions chosen lie within the range normally associated with natural ores.

Unless stated otherwise the synthetic leach solution contained 0.4 M  $\text{NH}_3/\text{NH}_4^+$  and 0.05 M  $\text{S}_2\text{O}_3^{2-}$ , at pH 9.5 with the addition of 10 mg/L Au and 5 mg/L Ag. The reagent concentrations were chosen based on prior work, thermodynamic analyses of gold leaching and speciation diagrams (Aylmore and Muir, 2001a, 2001b; Li et al., 1996; Wan, 1997). Fundamental aspects of gold and silver leaching with ammoniacal thiosulfate were also considered (Aylmore and Rae, 2001; Breuer and Jeffrey, 2000; Breuer et al., 2001; Jeffrey, 2001; Jeffrey et al., 2001a, 2001b). Stock solutions of reagents and precious metals were adjusted to pH 9.5. Experiments were performed by bottle rolling 600 mL solutions containing 400 g of mineral sorbents. A blank was also run to confirm that no loss occurred due to adsorption on the surface of the bottles. Sub-samples were taken at 0.5, 2 and 5 h. The experiments were carried out at ambient temperature regulated in the laboratory at around 15 °C. The experiments where possible were grouped to eliminate the impact of temperature variations on the test results.

The polythionate concentrations for experiments containing polythionates were determined by High Performance Liquid Chromatographic (HPLC) technique. Chromatographic separations of thiosulfate and polythionates were performed using an eluent of 0.005 M tetrabutyl-ammonium hydroxide, 0.005 M perchlorate and 0.005 M ammonia at a pH of 8.0. The HPLC equipment consisted of a Waters C18 Guard-Pak (5.0 × 6.0 mm ID) guard column and a Waters Nova Pak C18 column (150 × 3.9 mm ID) analytical chromatographic column connected to a diode array detector to measure the different solution species. Analytical grade sodium thiosulfate and sodium tetrathionate were obtained from BDH Pty Ltd. Sodium trithionate was synthesised from sodium thiosulfate and hydrogen peroxide following the method outlined by Kelly and Wood (1994).

Comparisons were made between the loss of reagents and stability of precious metals in solution for individual mineral components. Data from these experiments were used to quantify the extent to which mineral components 'preg-robbed' gold and silver contributed to the loss of reagents from solution. Duplicate experiments were conducted on many of the tests with only average results reported, due to the standard deviations of all the tests being within 3%. In all figures in this paper the gold and silver losses have been expressed as mg per kilogramme of mineral to gauge the adsorption capacity of some minerals under conditions typically used in leaching. Under the gold and

silver solution concentrations and mineral quantities used, a 100% loss of gold and silver in the presence of kaolinite or goethite is equivalent to 60 and 30 mg/kg<sub>mineral</sub> respectively. Whereas in the case for pyrite and activated carbon tests a 100% loss of gold and silver is equivalent to 300 and 150 mg/kg<sub>mineral</sub> respectively. In later tests where the silver concentration was doubled from 5 mg/L to 10 mg/L the values for gold apply.

## 3. Results and discussion

### 3.1. Role of mineral surfaces

#### 3.1.1. Effect of minerals on gold and silver adsorption

Equilibrium adsorption isotherms for gold and silver on pyrite, chalcopyrite and activated carbon in copper-free ammoniacal thiosulfate leach solutions are shown in Fig. 1. Each showed essentially linear plots of gold and silver loading versus equilibrium concentration and demonstrates significant preg-robbing capability. Activated carbon exhibited the highest preg-robbing ability since its specific surface area is significantly higher than other minerals (Table 1). However, based on surface area measurement, the gold loading per unit area is smaller (0.0001 mg/m<sup>2</sup>) than that observed for chalcopyrite (0.084 mg/m<sup>2</sup>) and pyrite (0.0093 mg/m<sup>2</sup>) in an ammoniacal thiosulfate solution containing 10 mg/L of gold.

In comparison, the sesquioxide minerals did not readily adsorb gold and silver under the conditions used. Since  $\text{Au}(\text{S}_2\text{O}_3)_2^-$  is a large linear anion, it has been considered that it would be difficult for this species to coordinate specifically with goethite surface hydroxyl groups (Machesky et al., 1991; Ran et al., 2002). A similar conclusion also holds for kaolinite.

#### 3.1.2. Effect of ammonia and thiosulfate concentrations on the adsorption of gold and silver

Fig. 2 shows the observed gold and silver concentrations lost to the minerals after bottle rolling in copper ammoniacal thiosulfate solutions for 2 h under different  $\text{NH}_3/\text{NH}_4^+$  and thiosulfate concentrations.

Generally the stability of gold and silver in solution was unaffected by reducing the ammonia concentration to 0.1 M in the presence of 0.05 M thiosulfate. In contrast Navarro et al. (2006) observed that adsorption of gold on the carbon increased with increasing ammonium hydroxide concentration up to 0.6 M and then remained constant, whilst the effect of ammonium hydroxide concentration was less significant at longer reaction times. They also observed that gold adsorption rate increased with increasing initial thiosulfate concentration, but decreased with extended length of time at higher thiosulfate concentrations.

In the presence of pyrite, significant gold and silver (>80%) reported to the mineral, and solution concentrations decreased when the ammonia concentration was below 0.4 M. Feng and van Deventer (2001) also found that preg-robbing of gold thiosulfate occurred on a range of sesquioxide and sulfide mineral surfaces when ammonia in solutions was below ~0.5 M – but in their case no free thiosulfate was present.

In the presence of pyrite, a thiosulfate concentration of around 0.05 M was required to reduce preg-robbing of gold and silver from 0.4 M  $\text{NH}_3/\text{NH}_4^+$  solution at pH 9.5. In contrast, results obtained by Feng and van Deventer (2002) indicated that in an oxygen-deficient system, a thiosulfate concentration of 0.5 M was required to eliminate preg-robbing completely in a solution containing 0.5 M ammonia. The lower levels of adsorption on goethite and kaolinite (~2% adsorption of gold and silver from solution) appear to be similarly affected by thiosulfate concentration – though the results with goethite need further clarification. Overall there was a limited reagent concentration range over which precious metals were stable in ammoniacal thiosulfate solutions at pH 9.5. To prevent precious metal loss onto sesquioxide minerals, reagent concentrations >0.2 M  $\text{NH}_4^+/\text{NH}_3$  and ≥0.05 M  $\text{S}_2\text{O}_3^{2-}$  are required. Pyrite requires ≥0.4 M  $\text{NH}_4^+/\text{NH}_3$  and ≥0.05 M  $\text{S}_2\text{O}_3^{2-}$  whilst activated carbon requires >0.1 M  $\text{NH}_4^+/\text{NH}_3$  and >0.05 M  $\text{S}_2\text{O}_3^{2-}$ .

**Table 1**  
Mineral components examined.

Mineral	Surface area (m <sup>2</sup> /g)	Quartz component (g)	Mineral component (g)	Percentage of mineral in mixture
Kaolinite	18.17	300	100	25%
Activated carbon	872	380	20	5%
Pyrite	3.53	380	20	5%
Chalcopyrite	0.39	380	20	5%
Goethite	13.3	300	100	25%

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