



Silver sulfide leaching with thiosulfate in the presence of additives Part I: Copper–ammonia leaching



Jared L. Deutsch*, David B. Dreisinger

Department of Materials Engineering, University of British Columbia, 6350 Stores Road, Vancouver, BC, Canada V6T 1Z4

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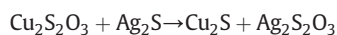
ABSTRACT

Thiosulfate is a promising alternative for the hydrometallurgical treatment of silver sulfide ores. In this study, the copper–ammonia catalyzed thiosulfate leaching of a silver sulfide rotating disk is investigated. The leaching of silver sulfide by a copper–ammonia thiosulfate solution occurs either by the substitution of cupric or cuprous for silver. The cupric reaction is favored due to a thermodynamic barrier to the cuprous reaction. At 1 mM copper, there is evidence of partial chemical control for copper–ammonia leaching with a maximum silver dissolution rate achieved when cupric is stabilized in solution with ammonia and for solutions with high thiosulfate availability. The addition of ethylenediaminetetraacetic acid decreased the leaching rate of the silver sulfide disk due to decreased cupric reactivity, although this comes with the benefit of increased cupric stability.

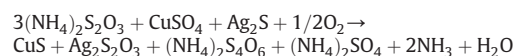
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1. Introduction

Concern about the environmental impact of cyanidation and the rising silver price has generated significant interest in alternative processes for the hydrometallurgical treatment of primary silver ores. These ores often contain large amounts of silver sulfide. Ores which may be amenable to gold cyanidation, such as the epithermal gold/silver deposit at the Peruvian Yanacocha mine, can suffer from limited silver extraction when process optimization is driven by gold recovery. Increasing cyanide concentration will increase silver extraction; however it may also increase mercury dissolution and environmental risks. Thiosulfate, a promising alternative to cyanide which has been historically used for silver leaching in the Patara process (Fleming, 2007), is receiving increased attention in the literature. This two part paper will focus on the application of thiosulfate to the leaching of silver sulfide with a number of additives. This system was first analyzed with significant detail by Flett et al. (1983) who proposed two reactions for silver sulfide leaching with cupric or cuprous. They claim that a cuprous substitution reaction will proceed in the absence of air forming chalcocite (Cu_2S):



In the presence of air, a cupric substitution reaction is proposed which forms covellite (CuS):



* Corresponding author. Tel.: +1 604 822 4805; fax: +1 604 822 3619.
E-mail address: jdeutsch@ualberta.ca (J.L. Deutsch).

In the first part of this study a fundamental hydrometallurgical investigation of copper–ammonia catalyzed thiosulfate leaching of silver sulfide is undertaken to investigate these reactions further. Using a rotating disk of synthetic silver sulfide, the effects of reagent concentrations, rotation speed, temperature and pH are each investigated separately. Of these variables, thiosulfate concentration, copper concentration and solution pH have the greatest effect on the silver sulfide leaching rate.

In the second part of this study, promising alternatives to the copper–ammonia system are tested including ferric ethylenediaminetetraacetic acid (EDTA), ferric oxalate and ferric citrate. The results of the rotating disk silver sulfide leaching studies are then applied to leaching a silver sulfide ore from Yanacocha.

2. Background and literature review

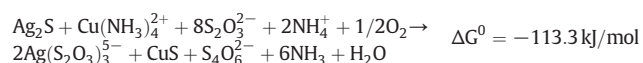
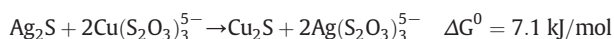
The cupric–ammonia catalyzed system is the most widely investigated thiosulfate system (Abbruzzese et al., 1995; Alonso-Gómez and Lapidus, 2009; Flett et al., 1983; Zipperian et al., 1988). This system can rapidly leach gold and silver (Molleman and Dreisinger, 2002); however thiosulfate quickly degrades to tetrathionate and trithionate in the presence of cupric species or oxygen (Ahern, 2005). For this reason, alternatives to the cupric–ammonia complex including ferric EDTA (Feng and van Deventer, 2010) and ferric oxalate (Chandra and Jeffrey, 2005) are being actively researched. These studies are almost all aimed at increasing metallic gold and silver recovery. There have been a number of studies on the leaching of silver-sulfide with thiosulfate solutions (Alonso-Gómez and Lapidus, 2009; Briones and Lapidus, 1998; Flett et al., 1983), but more work investigating the application of this promising system is necessary

to improve understanding of the complicated chemical and physical interactions.

2.1. Silver sulfide leaching with thiosulfate

Leaching of silver sulfide with the thiosulfate–ammonia–copper system has been previously studied by Flett et al. (1983), Briones and Lapidus (1998) and Alonso-Gómez and Lapidus (2009). There are a number of observations that can be gathered from these studies. Silver sulfide dissolution increased with increasing thiosulfate concentration and increasing copper concentration. Flett et al. (1983) found that the addition of ammonium hydroxide increased silver dissolution slightly while Briones and Lapidus (1998) found that increasing the total ammonium in the system decreased silver dissolution. This may be because silver dissolution was aided by the increased basicity from the addition of ammonium hydroxide in the research of Flett et al. while Briones and Lapidus maintained a constant pH by adjusting the amount of sodium hydroxide they added. Flett et al. also found that silver sulfide leaching with cuprous–thiosulfate (no ammonia) was possible, although the silver leached after 24 h was lower than it was with the addition of small amounts of ammonia. Alonso-Gómez and Lapidus (2009) took a different approach and used phosphate to inhibit lead leaching when EDTA is added to solution to increase metal recovery.

Using the most prevalent species in the cupric–ammonia–thiosulfate system based on speciation calculations (Aylmore and Muir, 2001), the reactions of Flett et al. (1983) can be rewritten. All of the reactions subsequently presented in this article are presented using either the metastable species of interest, or the most abundant species considering thermodynamic speciation with Pourbaix type diagrams as documented in numerous articles (Ahern, 2005; Alonso-Gómez and Lapidus, 2009; Aylmore and Muir, 2001). The reactions rewritten:

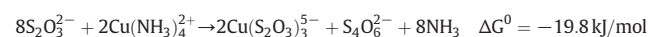
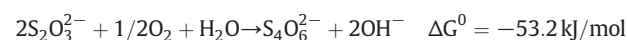


If cuprous reacts with the silver sulfide, then a molar ratio of 1 Cu^+ to 1 Ag^+ is required while if the cupric reaction proceeds a molar ratio of 1 Cu^{2+} to 2 Ag^+ is required. When the leach residues were analyzed with x-ray diffraction (XRD) (Briones and Lapidus, 1998; Cui et al., 2011; Flett et al., 1983), chalcocite and covellite were observed. However, in most of their experiments, a mixed mode is observed where the molar ratio of silver dissolved to copper precipitated varies from 1.1 to 1.9 depending on oxygen availability.

This copper precipitation phenomenon has also been observed by Briones and Lapidus (1998) and Cui et al. (2011). When leaching silver sulfide with a copper–thiosulfate solution (no ammonia), Cui et al. observed chalcocite (Cu_2S) formation with XRD due to the immediate reduction of cupric to cuprous in the absence of ammonia. Briones and Lapidus suggested that a mixed copper–sulfur phase such as digenite (Cu_{2-x}S) may have formed as leaching a stoichiometric amount of silver sulfide in a cupric–ammonia thiosulfate solution resulted in some leftover copper in solution. It has been suggested by multiple researchers that the leaching reaction may favor the formation of chalcocite (Cu_2S) due to the structural similarities between chalcocite and acanthite; however they are quick to point out that different diffusivities, reaction rates and ammonia complexes may exert a greater influence. In addition, the thermodynamic driving force for the cuprous reaction is less favorable compared to the favorable cupric reaction.

2.2. Loss of cupric complexes and thiosulfate

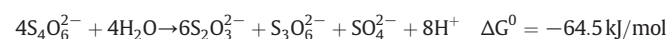
A significant challenge to using a cupric–ammonia catalyzed thiosulfate leaching system is the inherent instability. A neutral to alkaline pH must be used to prevent disproportionation of thiosulfate to sulfite and sulfur (Breuer and Jeffrey, 2003b). In an alkaline cupric–ammonia containing leach solution, thiosulfate oxidation will occur by either oxygen in solution or cupric:



Oxidation of thiosulfate in alkaline solutions by dissolved oxygen in solution is slow (Breuer, 2002); significant oxidation of thiosulfate is only observed in the presence of sulfur oxidizing bacteria or an oxidizing agent such as cupric. In the absence of any ammonia, the reduction of cupric in solution and oxidation of thiosulfate is nearly quantitative and complete in seconds (Breuer and Jeffrey, 2003b). In the presence of dissolved oxygen, cuprous species will be re-oxidized to cupric so a constant decrease in thiosulfate concentration is observed.

The primary oxidation product is tetrathionate ($\text{S}_4\text{O}_6^{2-}$), although other degradation products can be generated such as trithionate ($\text{S}_3\text{O}_6^{2-}$) and sulfite (SO_3^{2-}) (Ahern, 2005). These polythionates will eventually be oxidized to sulfate (Breuer and Jeffrey, 2003b). This reaction is catalyzed in solutions containing dissolved oxygen (Breuer and Jeffrey, 2003a). In addition, increasing the total amount of copper ions in solution increased the cupric:cuprous ratio and subsequent thiosulfate oxidation.

Tetrathionate may disproportionate to form thiosulfate and trithionate (Zhang et al., 2005). This process can result in the eventual regeneration of a small portion of oxidized thiosulfate and is catalyzed by bacteria such as *Thiomonas intermedia* K12 (Wentzien and Sand, 2004).



Further research on thiosulfate degradation in cupric–ammonia leach solutions has been undertaken by numerous researchers (Ahern, 2005; Aylmore and Muir, 2001; Breuer and Jeffrey, 2003a, 2003b; Jeffrey et al., 2003).

2.3. Cupric–ammonia thiosulfate leaching with the addition of EDTA

EDTA has been proposed as an additive to traditional cupric–ammonia thiosulfate solutions (Alonso-Gómez and Lapidus, 2009; Feng and van Deventer, 2010). EDTA strongly complexes a portion of the cupric in solution and reduces the amount of thiosulfate consumed during leaching by the reduction of cupric to cuprous. When leaching pure gold with a 0.1 M thiosulfate, 0.7 M total ammonia, 50 mg/L (0.8 mM) cupric solution, Feng and van Deventer found that adding 0.125 mM EDTA increased the amount of gold dissolved by approximately 25%. Adding more EDTA (up to 1.0 mM) greatly decreased gold dissolution rates. Although increasing EDTA additions decreased gold dissolution for the pure metal, when leaching a sulfide ore for 48 h under the same conditions, the addition of EDTA increased gold extraction. The improvement in total gold extraction showed an almost linear improvement with increasing EDTA additions of up to 2.0 mM, attributed to a decrease in thiosulfate consumption and prevention of surface passivation. This same improvement was also observed for silver extraction from the sulfide ore during leaching.

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