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Comparative study of auxiliary oxidants in cyanidation of silver sulfide

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ABSTRACT

Lower extraction of silver than gold in conventional cyanidation process has been commonly observed in practice. Acanthite (Ag_2S) is one of the typical silver minerals with low solubility in aqueous cyanide medium, which contributes to low overall silver extraction. The performance of different auxiliary oxidants, including hydrogen peroxide (H_2O_2), calcium peroxide (CaO_2), and LeachWELLTM, in cyanidation of synthetic silver sulfide has been investigated with the purpose of increasing the rate and extent of silver extraction. The addition of H_2O_2 does not only promote cyanidation of silver sulfide, but also increases the consumption of cyanide and hydroxide. Comparatively, the addition of CaO_2 exhibits less impact on silver extraction under the experimental conditions. LeachWELLTM has a better performance than H_2O_2 and CaO_2 in terms of increasing silver extraction and reducing cyanide and alkaline consumptions. The slurry pH exhibits insignificant effect on the performance of LeachWELLTM during leaching of silver sulfide. Similar performance of LeachWELLTM and lead nitrate on cyanidation of silver sulfide indicates that $Pb(NO_3)_2$ acts as the main active component in cyanidation process. Leaching of natural lead-bearing acanthite sample without auxiliary oxidant results in higher silver extraction compared to leaching of synthetic Ag_2S . Addition of H_2O_2 or CaO_2 during leaching of lead-bearing acanthite can promote silver extraction while the addition of LeachWELLTM exhibits little effect.

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1. Introduction

Silver is usually recovered along with gold from gold ore or concentrate through cyanidation which involves gold/silver dissolution in aerated cyanide solution to form gold and silver cyanide complexes. The Elsner equation reveals the stoichiometry of the reaction as the following (Marsden and House, 2006):

$$4Au/Ag + 8CN^{-} + O_2 + 2H_2O = 4Au(CN)_2^{-}/Ag(CN)_2^{-} + 4OH^{-}.$$
 (1)

However, the mineralogy difference between silver and gold leads to the variation of their final extractions (Xie and Dreisinger, 2009; Xie, 2006). Compared to gold, which almost always occurs in metallic form (Marsden and House, 2006), silver mineralogy is more complicated, for example, silver minerals such as silver sulfides (Ag₂S), argentojarosite (AgFe₃[SO₄]₂(OH)₆) and silver-containing manganese minerals, which usually yield low silver extraction in the conventional cyanide leaching process.

Abbreviation: LW, LeachWELL™.

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The dissolution of silver sulfide in the absence of oxygen in cyanide solution is governed by the following reaction (Xie and Dreisinger, 2007) (Speight, 2005):

$$Ag_2S + 4CN^- = 2Ag(CN)_2^- + S^{2-} Log K = -9.7.$$
 (2)

The equilibrium constant of the dissolution of silver sulfide in cyanide to form silver-cyanide complexes and sulfide anion is too low to guarantee satisfactory silver extraction. Therefore, the sulfide in acanthite needs to be oxidized during cyanidation to get better silver extraction. During cyanidation, sulfide ions may be oxidized in the presence of oxygen (Xie and Dreisinger, 2007);

$$2S^{2-} + 1/2O_2 + H_2O = S_2^{2-} + 2OH^-$$
(3)

$$S_x^{2-} + O_2 + 20H^- = S_2O_3^{2-} + S_{x-2}^{2-} + H_2O. \tag{4}$$

The stoichiometric relation of the leaching reaction during silver sulfide cyanidation has been investigated and a model to simulate the silver sulfide dissolution with different cyanide and oxygen concentrations has been established by Luna-Sanchez and Lapidus (2000). In their experiment, pure oxygen and 0.01 M NaCN solution were applied to dissolve pure silver





sulfide. The oxidation reaction was established as following (Speight, 2005):

$$\begin{array}{l} 2Ag_2S + 10CN^- + O_2 + 2H_2O = 4Ag(CN)_2^- + 2SCN^- + 4OH^- \\ \Delta G = -388.42 \ \ kJ/mol. \end{array} \tag{5}$$

The negative value of ΔG indicates that the reaction should be spontaneous from left to right, thus they believed that thermodynamically the dissolution of silver sulfide would proceed to the right in the presence of oxygen. There is the possibility that thiocyanate ion be further oxidized to form $S_2O_3^{-2}$. In fact, when pure oxygen and a 0.01 mol/L NaCN solution were applied in cyanidation of Ag_2S , they found that the dominant sulfide species in the cyanide solution was the $S_2O_3^{-2}$.

Therefore, the dissolution of silver sulfide in cyanide solution with the presence of only oxygen as oxidant maybe limited by the dissolution extent of Ag₂S in the cyanide solution and the extent of sulfide oxidation plus sequestering by cyanide. However, the low availability of oxygen in cyanide solution is considered as one of the key factors to hinder ideal silver extraction from acanthite cyanidation (Xie and Dreisinger, 2007).

To accelerate the leaching rate and improve the efficiency of the silver sulfide cyanidation process, Xie and Dreisinger (2007) investigated the addition of ferricyanide (Fe(CN) $^{3-}_{6}$) to acanthite cyanidation. Their conclusions indicated that ferricyanide was an effective oxidant for the cyanidation of gold/silver minerals. However, the use of ferricyanide in treating industrial materials, with mineral presented, would likely result in excessive consumption of ferricyanide.

Hydrogen peroxide (H_2O_2) has been proposed as an auxiliary oxidant in cyanidation of gold/silver ores (Arslan et al., 2003; Sun et al., 1993), by the reactions indicated in Reactions (6)–(9). The decomposition of H_2O_2 produces only oxygen and water, increasing the oxygen in the alkaline solution without introducing new impurities (Zhang, 1995). In addition, solubility in the cyanide solution is not a problem for H_2O_2 . Guzman et al. (1999) investigated the chemical and electrochemical process during gold cyanidation in the presence of H_2O_2 . It was shown that at concentrations lower than 0.01 mol/L, H_2O_2 did not oxidize free cyanide. Furthermore, research had been done to show that when H_2O_2 content is under 0.01 M, the leaching rate and final gold extraction increase with the H_2O_2 addition, so that H_2O_2 did not inhibit the cyanidation (Liu et al., 1998; Yang et al., 2003).

$$1/2O_2 + H_2O + 2e = 2OH^- \quad E^\circ = 0.4V (SHE)$$
(6)

$$H_2O_2 + 2e = 2OH^ E^\circ = 1.01V (SHE)$$
 (7)

$$H_2O_2 = H_2O + 1/2O_2 \tag{8}$$

$$2Au + 4NaCN + H_2O_2 = 2NaAu(CN)_2 + {}^22NaOH$$
 (9)

Calcium peroxide (CaO₂) has also been considered as an auxiliary oxidant for silver sulfide cyanidation. The application of CaO₂ as the replacement of H₂O₂ was first announced by a company named Interon in 1989 (Yang et al., 2003). Compared to H₂O₂ assisted gold leaching, the application of CaO₂ is much cheaper and more convenient (Sun et al., 1993). The mechanism is claimed to be similar to that of H₂O₂ in the cyanidation process. The only difference is that CaO₂ can react with alkaline solution first and generate H₂O₂ (Reaction (10)).

$$CaO_2 + 2H_2O = Ca(OH)_2 + H_2O_2$$
(10)

LeachWELL™ is a reagent grade catalyst formulated for fast cyanide leaching of gold, silver and other cyanide leachable metals. It was

| Fire assay and ICP analysis results of natural acanthite sample. | |
|--|--|

| Element | Ag | Total S | S ²⁻ | Fe | Pb | Cu | Zn |
|---------|-------|---------|-----------------|-------|-------|------|------|
| Wt.% | 33.20 | 26.15 | 24.64 | 18.10 | 11.38 | 0.16 | 0.03 |

Table 2

Leaching conditions of baseline experiment.

| Volume of solution | 1 L |
|--------------------|-----------------------------------|
| Solid | 0.1 g synthetic Ag ₂ S |
| Solution pH | 12 |
| Temperature | Room temperature, 17 °C (±3 °C) |
| Atmosphere | Air |
| Rotation speed | 500 rpm |
| Leaching time | 60 h |
| NaCN | 0.5 g/L |

initially developed by David Menne and marketed by an Australian company named Mineral Process Control. It is indicated by its supplier that when used with 5% cyanide, the dissolution rate of gold, copper and silver is increased more than sixty fold (MPC PTY LTD website, 2013, http://www.mineralprocesscontrol.com.au.). LeachWELL™ comes as small granules, about 4 to 6 mm in diameter, and it is reasonably soluble in cyanide solution. It consists of approximately 2% of lead nitrate (a typical additive to improve cyanide leaching) and 98% of a mild organic oxidant. This reagent is used along with intense leaching in analytical labs (Elder, 2000).

Lead nitrate $(Pb(NO_3)_2)$ is commonly used as an additive in gold cyanidation. $Pb(NO_3)_2$ is not believed to act as an oxidant and its main effect is believed to come from the function of lead ion in promoting leaching (Fink and Putnam, 1950; Jeffrey and Ritchie, 2000; Marsden and House, 2006). Jeffrey and Ritchie (2000) indicated that the main function of lead in the solution was to modify the surface of gold minerals by cementation to prevent the formation of insoluble layer that inhibits cyanide leaching (Reaction (11)).

$$Pb^{2+} + 2Au + 4CN^{-} = Pb + 2Au(CN)_{2}^{-}$$
(11)

Sandenbergh and Miller (2001) claimed that lead ion inhibited the formation of the passive layer by occupying sites on the surface, which prevented the cyanide from acting as a bidentate ligand necessary for the formation of the...-Au-CN-Au-CN-... polymer. Wu et al. (2002) indicated that lead ion in the solution was reduced and formed an alloy with metallic gold. This led to the formation of a galvanic cell, which accelerated the dissolution of gold.

In sulfide mineral cyanide leaching systems, Breuer et al. (2008) noticed that the oxidation of sulfide ion was quite slow. However, sulfide concentration decreased rapidly with the presence of lead ion. Dai and Jeffrey (2006) indicated that, when leaching sulfide minerals with cyanide, sulfide oxidation by oxygen was catalyzed in the presence of lead ion. They claimed that the role of lead was to promote the precipitation of PbS and enhance the anodic oxidation.

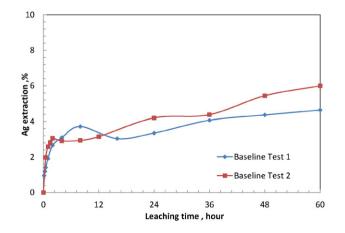


Fig. 1. Ag extraction vs leaching time in baseline experiments (0.5 g/L NaCN, pH = 12, room temperature, 500 rpm).

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