Minerals Engineering 125 (2018) 15–25

Contents lists available at ScienceDirect

Minerals Engineering

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

Effect of concurrent ball milling on cementation reactions: The case of Cu-Al system

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ARTICLE INFO

Keywords:
Copper cementation
Aluminum powder
Aluminum scrap
Ball milling
Central composite design
Reaction mechanism
Activation energy
Deposit morphology

ABSTRACT

Despite a strong thermodynamic tendency, cementation by aluminum of relatively noble metals from sulfate solutions involves a rather slow kinetics due to the presence of the passive oxide film on the aluminum surface. We know from the literature that small amounts of chloride ion can reduce the dimension of the problem. In this paper, we investigate the additional effect of concurrent ball milling on the enhancement of the cementation kinetics of Cu-Al system as a case study. The cementation tests were carried out in a polyamide jar placed in a planetary ball mill. In the first part of the study, the effect of rotation speed of the milling jar, number of alumina milling balls, particle size of the aluminum powder, and the initial hydrogen ion concentration on the copper recovery was studied and optimized using a central composite design of experiments. In the second part, we investigated the kinetics and mechanism of the cementation process under the optimum conditions obtained from the first part. The results show that the concurrent ball milling can shorten or even remove the initial slow rate period, known as the induction period. It also alters the rate-controlling mechanism from chemical to boundary layer diffusion revealed by a decrease in the activation energy from 79 to 18 kJ mol^{-1}. The morphology of copper deposits obtained under ball milling condition was found to be botryoidal in contrast to the typical dendritic structure. The findings overall support the beneficial role of the simultaneous ball milling in lowering the kinetic barrier of cementation reactions on the aluminum surface.

1. Introduction

Cementation is probably the oldest hydrometallurgical process that involves reductive precipitation of a metal ion from a solution by a more active metal (Lung, 1986). Having a large negative reduction potential, aluminum has served the precipitant metal of choice for different cementation systems such as gold (Wang et al., 2007; Karavasteva, 2010), silver (Karavasteva, 2009; Mackinnon and Ingraham, 1971a), lead (Farahmand et al., 2009), mercury (Anacleto and Carvalho, 1996), platinum and palladium (Kim et al., 2010), and most of all, copper (Anastassakis et al., 2015; Annamalai et al., 1978a; Annamalai et al., 1978b; Lin'kov and Temiraev, 2009; Mackinnon and Ingraham, 1970; Mackinnon and Ingraham, 1971b). The usage of aluminum in those systems has some advantages over other commonly used active metals. For example, for the copper cementation process, aluminum offers the following benefits over iron:

1. The theoretical consumption of aluminum is only 32% of iron by weight comparing their molar masses (27 and 56 g mol^{-1}, respectively) as well as their oxidation states relevant stoichiometric coefficients in the cell reactions (1) and (2) (2/3 and 1, respectively). This will compensate to a large degree the higher unit price of scrap aluminum over scrap iron.

$$\mathrm{Cu^{2+} + 2/3Al = Cu + 2/3Al^{3+}} \quad E^{\circ} = +2.00\ \mathrm{V} \tag{1}$$

$$\mathrm{Cu^{2+} + Fe = Cu + Fe^{2+}} \quad E^{\circ} = +0.78\ \mathrm{V} \tag{2}$$

2. Aluminum has only one oxidation state whereas iron has two. This causes unwanted side reactions for iron to take place. In air or an oxygen atmosphere, there is a slight decrease in the cementation efficiency as a result of the partial oxidation of $\mathrm{Fe^{2+}}$ ions to $\mathrm{Fe^{3+}}$, Eq. (3), followed by the reverse transformation consuming the cemented copper, Eq. (4):

$$\mathrm{2Fe^{2+} + 1/2O_2 + H_2O = 2Fe^{3+} + 2OH^{-}} \quad E^{\circ} = +0.458\ \mathrm{V} \tag{3}$$

$$\mathrm{Cu + 2Fe^{3+} = Cu^{2+} + 2Fe^{2+}} \quad E^{\circ} = +0.434\ \mathrm{V} \tag{4}$$

3. By using aluminum as the reductant, a useful solution of aluminum

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https://doi.org/10.1016/j.mineng.2018.05.024
Received 23 December 2017; Received in revised form 24 April 2018; Accepted 16 May 2018
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Table 1
Chemical composition of the aluminum powder.

Element	Al	Si	Cu	Zn	Other
wt%	99.48	0.20	0.14	0.10	0.08

salt (sulfate, chloride, etc.) is obtained that can be crystallized as a valuable by-product.

4. Vast amounts of aluminum swarf generated during machining and manufacturing of aluminum parts with a high surface area can effectively be utilized in the cementation process.

However, a kinetic barrier on the aluminum surface, the adherent oxide film that protects it from environmental corrosion, causes aluminum to exhibit a slow reaction rate in both the main cementation reaction and side reactions, such as evolution of hydrogen gas:

$$3H^+ + Al = 3/2H_2 + Al^{3+} \qquad E^\circ = +1.66\ V \tag{5}$$

Karavasteva (2005) has compared cementation of copper from sulfate solution by different fine metal powders (Zn, Fe, and Al) under similar conditions. She indicated that despite having a more active potential, Al reduced copper much slower than Zn and Fe because of the passive layer on its surface; such that Zn and Fe cemented respectively 99% and 98% of copper in 8 min, while the efficiency was only 25% in the case of Al.

Mackinnon and Ingraham (1970, 1971b) were the first who reported that the addition of small amounts of chloride ion can activate the aluminum surface by dissolving its protective oxide film. They demonstrated that the addition of only 10 mg L^{-1} of a chloride salt would increase the rate constant of the Cu-Al cementation reaction up to 5 times and alter the rate-controlling step from chemical reaction to diffusion through the boundary layer. Being small, mobile, and active, chloride ions easily adsorb on the oxide film of the aluminum surface. The result is the dissolution of aluminum just like the pitting corrosion (Godard and Torrible, 1970).

There are ample information on the pitting corrosion of aluminum by chloride ion. It is known that the surface of aluminum in aqueous solutions is usually composed of an oxyhydroxide type material such as AlOOH. It has a positive charge at pH values less than 9 (the isoelectric point of Al in aqueous solutions) (Natishan and O'Grady, 2014). The presence of this attractive force allows Cl^- ions to readily move towards the oxide/solution interface and interact with the passive film by different mechanisms proposed by McCafferty (2003). The interactions may eventually lead to the generation of soluble aluminum hydroxylchloro complexes of the form $Al(OH)_2Cl_2^-$ according to the plausible following reactions:

$$Al_2O_3 + H_2O = 2AlOOH \tag{6}$$

$$AlOOH + H^+ + 2Cl^- = [Al(OH)_2Cl_2]^- \tag{7}$$

Kolics et al. (1998) have shown that Cl^- adsorption is pH dependent, with the Cl^- concentration on the surface decreasing with increasing pH. This result supports the reaction scheme given above.

In several papers, Annamalai et al. (1978a, 1978b), Annamalai and Hiskey (1978), Annamalai and Murr (1978), Murr and Annamalai (1978) have reported their findings on the Cu-Al cementation system with rotating disc geometry. They also used chloride ion in their experiments to dissolve the oxide film on the aluminum surface. They examined the effects of the quantity and the source of chloride ion as well as Cu^{2+} concentration, pH, temperature, rotation speed and ultrasonic radiation on the reaction kinetics and morphology of the cemented copper. They suggested at least 75 mg L^{-1} chloride ion in the copper sulfate solution to activate the aluminum passive surface.

Kanungo et al. (2001) used perchlorate ion (ClO_4^-) to dissolve the aluminum oxide film in the immersion plating of copper onto aluminum, a process that is based on cementation reaction. Chen and Lee (1994) investigated the effect of different chelating agents on removing the oxide film in the presence of chloride. In addition, Dönmez et al. (1999) and Ekmekyapar et al. (2012) studied the Cu-Al cementation kinetics using Levich equation to determine the diffusion coefficient. In contrary to the above-mentioned studies that were performed in acidic media, Djokić (1996) investigated cementation of copper onto aluminum in NaOH solutions. He showed that despite the easy dissolution of the aluminum oxide film in the solution, the reaction rate is still low because of the high stability of alkaline copper complexes as well as high hydrogen evolution.

In addition to the usage of chloride ions, the problem of the surface passivity of aluminum may be tackled by conducting the cementation reaction under ball milling conditions. With this idea in mind, we devised a ball milling-assisted cementation set-up to explore the effectiveness of the intended process.

The conventional experimentation methodology of the so-called "one-factor-at-a-time" employed by most researchers is incapable of showing any interactions between the process variables, and therefore, do not lead to the real optimum condition. An alternative and better methodology that gives the most amount of information out of a given expenditure of experimental effort is statistical "design of experiments" (DOE). It is based on Fisher's factorial concept by testing all possible combinations of factor levels (Montgomery, 2013). Few cementation systems have been investigated by adopting DOE methodology including cementation by iron of copper and silver reported by Djoudi et al. (2007) and Yahiaoui et al. (2010), respectively, and cementation of copper by aluminum reported by Lin'kov and Temiraev (2009).

In the present paper, we report the results of our investigation concerning the effect of concurrent ball milling on the cementation of copper by aluminum powder with the aim of improving the efficiency and kinetics of the cementation reaction. The research consisted of two consecutive parts. In the first part, we adopted a statistical experimental design approach to identify the most influential factors and optimize the recovery of copper by a response surface methodology (RSM). In the second part, we performed experiments at the optimum conditions obtained from the first part to explore the kinetics and mechanism of the adopted process. We chose Cu-Al cementation system as the case study because of the vast availability of the kinetic data on copper cementation that makes comparison possible between the results of different studies.

Table 2
Levels and values of the factors.

Factors	Levels				
	−2	−1	0	+1	+2
X_1: RS (rpm)	50	100	150	200	250
X_2: BALL (#)	0	1	2	3	4
X_3: PS (μm)	279	754	1229	1704	2179
X_4: H (10^{-3} mol L^{-1})	0.06 (pH = 4.22)	0.53 (pH = 3.28)	1.00 (pH = 3.00)	1.47 (pH = 2.83)	1.94 (pH = 2.71)

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