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Leaching of silver from solid waste using ultrasound assisted thiourea method

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Abstract

Thiourea leaching of precious metals such as gold and silver from ores has several advantages when compared with conventional cyanidation process. In recent years, the use of ultrasound in leaching processes is becoming increasingly popular in hydrometallurgy. This paper deals with combining these two techniques for silver leaching from solid waste of a cyanidation leach plant located in Kütahya, Turkey. The primary aim of this research is to assess the technical performance of the method. To achieve maximum leaching yield, eight process variables have been selected to estimate optimum process conditions by means of statistical factorial design and steepest ascent techniques. Laboratory-scale experiments showed that complete leaching of silver may be achieved by this process.

Keywords: Silver; Thiourea; Ultrasound; Leaching; Optimization; Steepest ascent

1. Introduction

Silver is one of the widely used metals in industry. The majority of silver is consumed in film processing industry with 40–50% of overall consumption, electrical industry follows with 20–30%, and ornaments and jewelry consist of 10% of overall consumption. In addition to the recovery of silver from scrap metals, silver is produced either directly from ores or as a side product during the productions of zinc, copper and lead.

Today, silver production is mainly performed by the conventional cyanidation process. Thiourea (NH_2-$CSNH_2$), an alternative non polluting reagent for extracting precious metals, has shown promise for implementation in the metallurgical industry [1]. Laboratory testing has indicated that thiourea process for gold and silver extraction has several advantages over the conventional cyanidation; greater selectivity towards gold and silver, fast extraction kinetics, low environmental impact, and easier handling of reagent [2–4]. An additional advantage over cyanide exists when treating refractory sulphide ores by bio-oxidative pretreatment in highly acidic solutions; a neutralization prior to cyanidation is required, while thiourea leaching occurs in acidic solution that can be treated directly [5,6].

The use of ultrasound for ore leaching becomes increasingly popular in hydrometallurgy. Sonochemical extraction techniques together with classical methods gave a faster [7,8] and selective [9] extraction of metals. Orlov [10] and Chizhikov et al. [11] used ultrasound for Cu leaching from Copper ores in sulfuric acid. The application of ultrasound in extractive metallurgy has been reviewed by Polyukhin [12]. The effect of ultrasound on ammonium leaching of zinc from Galmei ore has been investigated by Slaczka [13]. The use of ultrasound in nickel extraction from lateritic nickel ore using a strain of Aspergillus niger was studied by Swamy et al. [14]. The kinetics of the ultrasound assisted dissolution of phosphate rock in HNO_3 is modeled by Tekin et al. [15], and in HCl by Tekin [16].

Recently the effects of ultrasound for improving chemical reactions have been reviewed [17]. In solid–liquid systems, Ultrasound increase the rate of

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Nomenclature

S	step size in steepest ascent experiments	Y	process response (leaching yield)
X	coded value of the process variable	Z	absolute value of the process variable

dissolution principally by the *cavitation* effect leading to the appearance of many microcracks on the solid surface subjected to ultrasound. Ultrasound increases also the diffusion speed of soluble species in the liquid phase; this effect facilitates the leaching agents to reach more easily the bottom of capillaries [18]. Furthermore, if the solid reagent is in the form of a powder, ultrasound energy can cause particle rupture, with a consequent increase in surface area available for reaction. One might expect that the increase in surface area alone would be sufficient to explain any enhanced reactivity due to ultrasound [19].

The purpose of this work is to investigate the effect of ultrasonic energy on the leaching yield of silver content of a mining waste, and maximize the yield by means of response surface methodology. The effect of the ultrasound on the leaching kinetics and mechanism is planned to be investigated in a further study.

2. Experimental

2.1. Experimental planning

A significant part of the industrial experimentation is devoted to finding functional relationship between independent variables and process response:

$$Y = f(X_1, \dots, X_n) \tag{1}$$

Usually, at the beginning, a theoretical model is not of primordial importance, and an empirical one is sufficient for the final goal to estimate the optimum conditions by means of laboratory-scale experimentation. Efficient response surface methods have been developed and successfully used for this purpose [19]. If the experimenter has no prior information about the optimum conditions, the most widely used method to reach the proximity of the optimum point with minimum number of runs, is the classical *steepest ascent* (*descent*) technique.

The gradient of the model equation (1) gives the steepest ascent direction, which may be estimated locally by means of first-order linear approximate:

$$Y = b_0 + \sum_{i=1}^{n} b_i X_i \tag{2}$$

2^n full or fractional factorial design, where each variable runs at two levels, is widely used to obtain experimentally linear models [20]. The model parameters, b_i, are used to locate the steepest ascent path by the following relation:

$$\Delta X_i = S * b_i, \quad i = 1, \dots, n \tag{3}$$

where S is the step size adjusted by the experimenter. Starting from a suitable base point, X_b, coded values of the process variables, X_i; for a new experiment is calculated using relation (3) as follows:

$$X_i = X_{b,i} + \Delta X_i, \quad i = 1, \dots, n \tag{4}$$

Absolute value (Z_i) of the variables are calculated by the following relation;

$$Z_i = Z_{i,0+} X_i \cdot (Z_{i,2} - Z_{i,1})/2 \tag{5}$$

where $Z_{i,1}$ is low level, $Z_{i,2}$ is high level and $Z_{i,0}$ is medium level of the variable.

Thus, a *line search* along this path is conducted until no additional increase in the response, Y, is evident. In summary, the experimentation starts from an initial estimate of the optimum point, proceeds sequentially via gradient determination (factorial design) blocks and line search blocks, until to reach finally feasible optimum conditions.

2.2. Materials and method

Solid waste samples used in this study were collected from a silver ore processing plant (Gümüşköy 100 Yıl) located in Kütahya, where approximately 4000 tones of silver ores is processed by cyanide leaching method to recover approximately 122 tones of 1000 carat (99.99% in purity) silver per year. The plant started production in 1987 and planned to have 20 years economical life, at the end of which, approximately 25 million tones of solid waste will be stored in four different pools located within the factory.

$Ag_2Fe_5S_8$, Ag_2S and Ag_3SbS_3 are the principal silver minerals occuring in the raw ore [21]. Two types of ore exist in Gümüşköy area; one is the residue of an antique lead processing plant, with 2.5 million tons reserve and mean silver content of 374 ppm, the other has 16.7 million tons reserve with 167 ppm silver content.

Wet solid waste was dried at room temperature (approximately 20 °C) until constant weight. Samples were withdrawn from the air-dried waste, for sieve, chemical and X-ray analysis. The chemical composition of the air dried is given in Table 1: Principal components are detected by X-Ray F analysis (Rigaku RIX 1000), and Ag is detected by AAS Graphite Furnace (Perkin Elmer 6000 Model).

The schematic illustration of the experimental set-up is shown in Fig. 1. The leaching of the mining waste was carried out under atmospheric pressure, in a cylindrical