



Experimental process analysis and mathematical modeling for selective gold leaching from slag through wet chlorination



Antoneta Filcenco Olteanu^a, Tanase Dobre^b, Eugenia Panturu^a, Aura Daniela Radu^a, Ata Akcil^{c,*}

^a Research and Development National Institute for Metals and Radioactive Resources, 70 Carol I, Bvd., District 2, 020917 Bucharest, Romania

^b University Politehnica of Bucharest, 1-7 Polizu Street, District 1, 011061 Bucharest, Romania

^c Department of Mining Engineering, Mineral Processing Division (Mineral-Metal Recovery and Recycling Research Group), Suleyman Demirel University, TR32260 Isparta, Turkey

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ABSTRACT

The aim of this paper consists in defining optimal conditions of controlled and selective gold leaching contained in copper–gold slag, by wet chlorination. Studies were conducted on three types of copper–gold slag, resulting as by-products from pyrometallurgical processing of non-ferrous raw materials. Due to high levels of copper in slag, an oxidative pre-treatment is necessary to separate gold from accompanying elements and also for copper recovery. After nitric acid pre-treatment, nearly 87% of Cu was readily extracted, and lead and silver contents in all three types of solid samples have been reduced by 91.34% and 96.29%, respectively. Selective leaching of the solid phase resulting after HNO₃ pre-treatment was done by wet chlorination, using nascent chlorine (Cl₂) as the leaching agent, with the presence of hydrochloric acid (HCl). The chlorine was obtained “in situ” by the reaction of sodium hypochlorite (NaOCl, 5% Cl₂) and HCl. In this study, the effect of nitric acid pre-treatment, redox potential (Eh) given by sodium hypochlorite (NaOCl) consumption, hydrochloric acid concentration, temperature and leaching time have been studied. The potential required for a rapid leaching rate and for avoiding the reduction of gold is 950 mV (vs AgCl/Ag_(KCl sat)). The 5% NaOCl consumption required for gold leaching from pre-treated samples was lower than NaOCl consumption necessary for original sample leaching. The highest gold extraction yield (98%) was obtained after 6 h at ambient temperature, solid/liquid ratio (w/v) 1:2 and 4 M HCl. In order to explain experimental results a mathematical model of the simultaneous gold, copper, lead and silver dissolution has been developed. It considers at particle level, the competition between surface reaction and the diffusion of the active species by a gel coating that covers the dissolving particle.

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1. Introduction

The problem of gold recycling from secondary resources is the center of interest in all economies of the world, due to its vast industrial applications, high market prices and extensively used precious metal. Therefore, this problem not only is technological, legislative or economical, but also is related to the environmental protection (Ficeriová and Baláž, 2010). Depletion of rich resources of precious metals and the need for treatment of low content gold ores or with a complex mineralization, as well as the emergence of very strict environmental regulations have encouraged researchers to find alternative to cyanide extraction systems. These technologies must be competitive in order to compensate low cost cyanide, and should have salient features like selectivity, high recovery, economical and eco-friendly even when operating under small-scale conditions (Sayed, 2006). For these reasons,

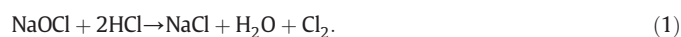
nowadays in the gold industry there are three main trends: elimination and even prohibition of cyanide extraction in gold mining; development of new technology for gold extraction; and attracting new unconventional reserves of raw materials.

Hydrometallurgical processes for gold recovery from secondary sources generally involve three stages: pre-treatment, recovery and refining of metals from aqueous solution. (Tuncuk et al., 2012) Leaching is commonly done by using a variety of chemicals which include nitric acid (Niederkorn and Huszar, 1984), mixtures of nitric, hydrochloric and sulphuric acids (Krupkova et al., 1987), mixtures of sulphuric acid, nitric acid and hydrogen peroxide (Kristofova and Karnik, 1996), aqua-regia (Lee et al., 1997), ferric chloride (Hanna, 1995), thiourea (Becker et al., 1983; Sheng and Etsell, 1988), potassium iso-cyanate (Khalemskii et al., 1998), potassium iodate and iodine (Leibovitz, 1998), iodide–nitrite mixture (Novoselov and Peshchevitskii, 1998), thiosulphate (Chandra and Jeffrey, 2005; Feng and Van Deventer, 2006; Navarro et al., 2002; Senanayake, 2004; Zhao et al., 1999) and cyanide (Leao and Ciminelli, 2000; Marsden and House, 2006).

* Corresponding author. Tel.: +90 246 2111321; fax: +90 246 2370859.
E-mail address: ataakcil@sdu.edu.tr (A. Akcil).

Conventional cyanidation is considered as the mainstream method for the leaching of gold and other precious metals from the complex ores and has been practiced over the years. The advantages of cyanidation process over other extraction methods include its simplicity, fast leaching kinetics, and ease of recovery. But due to the toxicity of the cyanide, its use in this process has been prohibited in many countries (Biswas et al., 2010). A suitable replacement for cyanide may be the use of thiosulphate (Ficeriová et al., 2005) due to the fact that thiosulphate is substantially less expensive than cyanide, and it facilitates the leaching of complex materials through matrix degradation (Grosse et al., 2003). Thiosulphate leaching to recover gold from ores is known for several decades but extensive researches have been started only for the past two decades (Zipperian et al., 1988). Thiosulphate leaching has many advantages over the cyanidation process including higher leaching rates and less interference from foreign ions (Tripathi et al., 2012). Another potential candidate to be used as lixiviant is thiourea (Baláz et al., 1996) because thiourea in acidic media can readily dissolve precious metals (e.g. gold) as a stable complex (Whitehead et al., 2009), which eventually facilitates leaching of metals from complex ores, concentrates and other potential sources of precious metals. Thiourea is a non-cyanide lixiviant for gold leaching which efficiently recovers gold (Orgul and Atalay, 2002) but this is not an economical route for the gold leaching due to its high consumption and cost. Bio-hydrometallurgy is one of the most promising new cleaner eco-friendly technologies. Bioleaching is a specialized bio-hydrometallurgical process. In this process either metabolic activities or products of microorganisms are involved. This process is based on the ability of micro-organisms (bacteria and fungi) to transform solid compounds into soluble and extractable elements, which can be recovered (Akcil and Deveci, 2010; Ciftci and Akcil, 2010; Ehrlich, 2004; Anjum et al., 2012). Halide systems such as bromine–bromide, iodine–iodide and bromide–chloride are capable of dissolving gold at very fast rates. These systems are strongly oxidizing and dissolution rates are typically several orders of magnitude faster than those achieved with cyanide and oxygen under ambient conditions (Hiskey and Atluri, 1988). Because of cyanide toxicity and environmental concerns, there is an increasing interest in finding alternatives leaching system for gold recovery. The chlorine leaching in aqueous media has been the first process used in gold hydrometallurgy since 1848 but was replaced by the more economical cyanidation process. The halogen/halide system predates cyanidation for the treatment of ores containing fine gold particles and gold sulfides ores not amenable to treatment by gravity concentration and amalgamation. The method lost popularity with the discovery of cyanidation, which proved to be cheaper and technically easier for the common miner. However, there has been a renewed interest in chlorination because of recent negative experiences with cyanidation and the potential environmental consequences of mercury use (Laatikainen and Paatero, 2005; Vieira, 2005; Tuncuk et al., 2012).

Chlorine leaching has few advantages due to the higher dissolution rate of gold, low price of leaching reagents and non-polluting character (Filcenca Olteanu et al., 2010; Putman, 2004; Radulescu and Filip, 2004; Radulescu et al., 2006; Welham and Kelsall, 2000). Dissolution of metallic gold was studied (Mahkatsi, 2006; Viñals et al., 1995) by generating chlorine “in situ” from the reaction between commercial sodium hypochlorite and hydrochloric acid (Eq. (1)):



The dissolution reaction occurs as follows (Baghalha, 2007) (Eq. (2)):



The ‘iGoli Mercury-free Gold Extraction Process’ was invented during the last few years by the Small Scale Mining and Beneficiation Division (SSMB) of Mintek in South Africa. The iGoli process is designed to leach gold from >0.1% gold concentrate to produce 99.9% gold product. The iGoli process is a modern version of chlorination, and uses a mixture of pool acid (dilute hydrochloric acid), bleach (sodium hypochlorite) and sodium metabisulphate to leach and recover metallic gold (Mahkatsi and Guest, 2003).

Hence the purpose of this study is to investigate a hydrometallurgical method to recover gold from copper–gold slag, resulting as by-products from pyrometallurgical processing of non-ferrous raw materials by wet chlorination. In this study, the effect of nitric acid pre-treatment, redox potential (Eh) given by sodium hypochlorite (NaOCl) consumption, hydrochloric acid concentration, temperature, leaching time and solid/liquid ratio have been studied.

2. Materials and methods

2.1. Reagents

All chemicals and reagents used were of commercial grade. Solution of nitric acid (HNO₃) with 32.5% mass concentration from Chimpar S.A Romania, solution of hydrochloric acid (HCl) with 35% mass concentration Chimpar S.A and sodium hypochlorite (NaOCl) solution with 5% chloride concentration from S.C. SUN Industries have been used.

2.2. Samples

Studies were developed on three types of copper–gold slag (sample codes: A, B, C) resulting as by-products from pyrometallurgical processing of non-ferrous raw materials from Transgold Baia Mare, Romania. The copper–gold slags were processed by disintegration followed by other chemical or physico-chemical transformations, which significantly affected the subsequent slag’s processing operations. First of all, the slag was crushed and sieved three times. First two crushing steps were performed using a jaw crusher (BB 200 Retsch). Preliminary crushing has reduced slag size from 200 mm to max. 60 mm and in the second step slag size was reduced from 60 to 20 mm. The last step of mechanical processing, before concentration, was performed in a laboratory installation consists of six ball mill PM 100CM series. After milling we obtained a powder with an average particle size of about 0.15 mm, optimum size for gold leaching. In order to obtain copper–gold hydro-gravitational concentrate a Gemini table (GT60) was used, recovering thus 92.87% of gold in concentrate, 3.28% in middling and 3.85% in tailing.

The microstructure and compositional microanalysis of the processed powders were characterized using a QUANTA INSPECT F scanning electron microscope (SEM-EDX). Samples were prepared by dispersing dry powder on double disked conductive adhesive tape. Samples were coated with carbon by arc discharge method for SEM-EDX. The chemical compositions of the processed slag were determined by the atomic absorption spectrophotometer (VARIAN spectral AA-88) and the silica content was determined gravimetrically.

2.3. Leaching tests

Oxidative pre-treatment of 5 kg copper gold hydro-gravitational concentrate (A, B, C) was carried out in a glass reactor (Duran) of 30 L capacity with a propeller agitator and integral heating/cooling system. The leaching tests were conducted at atmospheric pressure and ambient temperature, using 32.5% HNO₃ with solid–liquid ratio (w/v) 1:4 for 6 h. Stirrer speed was set to 700 rpm in order to keep slurry completely suspended. At the end of the oxidative pre-treatment the slurry was then filtered using water suction pump, through Whatman filter paper and washed thoroughly with cold water at solid–liquid

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