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Hydrometallurgical Recovery of Gold from Mining Wastes

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Abstract

Gold is a highly required material for a wide range of personal and industrial applications. The high demand for gold, together with the shortage of natural resources and high pollution potential of wastes generated during mining and ore processing activities led to search for alternative sources of gold. A possible source is represented by mine wastes resulting from the processing of polymetallic or sulfidic ores. The reprocessing of wastes and old tailings with moderate to low content of gold offers not only a business opportunity, but also enhances the quality of the surrounding environment, changes the land use and offers a wide range of socio-economic benefits. Cyanidation, the most widespread Au leaching option, is progressively abandoned due to the high risk associated with its use and to the low public acceptance. Therefore, alternative methods such as thiocyanate, thiourea, thiosulphate and halide leaching gained more and more interest. This chapter presents the most important features of some Au leaching methods, emphasizing their advantages, limitations and potential applications.

Keywords: gold recovery, nonferrous mining wastes, leaching, technology, circular economy

1. Introduction

From ancient times, gold was associated with power and wealth, being used for the manufacture of tools and weapons, decorative objects and jewelries. Nowadays, gold is a highly required element in the field of electronics, nanotechnology, medicine, food, cosmetics, decorative or creative fashion, as well as in space technology [1–4].

Despite their economic importance, mining and ore processing have a low public acceptance and a high environmental pollution potential as they generate great amounts of wastes consisting in a mixture of solid waste materials (sand, fine grained ground-up rock with a size of 1–600 μm), water, chemicals and high concentrations of hazardous metals (Cu, Cd, Fe, Mn, Pb, Zn), metalloids (As, Se), together with precious metals (Au, Ag) [5–9].

The mining waste disposal and the long-term management of tailing storage facilities are critical issues of the mining process, as they can cause landscape damages and can lead to severe environmental contamination and destruction of living

ecosystems [10, 11]. Worldwide, several tailing dam accidents with significant environmental damages and sometimes also with human lives loss were reported [11, 12]. One of the most important tailings dam failures occurred in 2000 in Baia Mare, Romania, due to an improper design and lack of effective management of the tailings impoundment, causing catastrophic damages to the environment [13, 14]. Another severe mining failure was reported in 1971 in the Certej River catchment, Romania, where a large volume of mine tailings flooded the valley and generated a significant pollution of the surrounding environment, together with 100 human lives loss [15]. An important pollution accident occurred in Spain in 1998, when a massive amount of acid water and mud containing toxic metals were released into the Agrio and Guadiamar River from the dam of the Aznalcollar tailings pond [16].

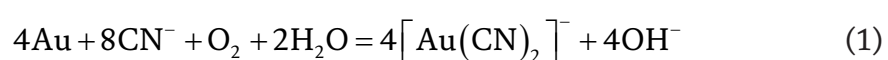
Mining wastes and by-products are valuable secondary resources since they contain important amounts of base, precious or strategic metals that can be recovered [17]. Beside their economic importance, the wastes valorization by reprocessing could improve the quality of the environment. The recycling, reuse and recovery of extractive wastes is also encouraged in the Extractive Waste Directive 2006/21/EC as it could lead to sustainable mining by resources conservation and reduced environmental impact [17, 18]. Therefore, new processing methods and technologies need to be developed and implemented. The most used method for Au leaching from wastes is based on cyanidation. However, in the context of raising social awareness and straightening environmental regulation, cyanide has become a socially and environmentally undesirable method for Au recovery. Therefore, the interest in finding other alternatives to cyanide, with low impact on the environment increased [19–22]. There are a considerable number of gold leaching methods which are currently tested or developed as pilot-scale studies, but only a few are available on commercial scale. The most tested leaching agents for gold are thiosulfate, thiourea, thiocyanate and halides [19–21]. An effective Au recovery process from mining wastes could provide new resources for various industrial fields [23]. This chapter presents an overview of the most used leaching methods for Au recovery with their advantages and limitations.

2. Gold leaching methods

2.1 Cyanide leaching

Cyanide is a cheap but highly toxic reagent that is very effective in leaching gold from low-grade minerals and mining wastes. Therefore, cyanidation is the most widely used method for the extraction of gold from mining wastes. Despite its advantages, is considered an unacceptable and highly hazardous approach. The health and safety concerns related to the use of cyanide as a leaching agent were raised after the occurrence of several technological accidents that severely damaged the environment [20, 24, 25]. Such incidents led to the development of a voluntary program in the gold mining industry entitled the “International Cyanide Management Code” with the objective to safely manage cyanide and safeguard human health and the environment [26, 27].

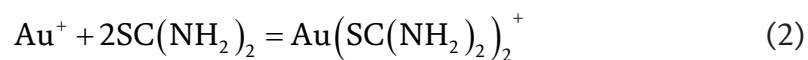
The cyanide leaching is based on the Au complexation with cyanide ions in the presence of oxygen in an alkaline solution according to Eq. (1) [22].



Cyanide compounds are classified into three main types: free cyanides (hydrogen cyanide (HCN), ionic cyanide (CN⁻)), weak acid dissociable (WAD) cyanide complexes (with Cd, Cu, Ni, and Zn) and strong cyanide complexes (with Co, Au, Ag, and Fe). The chemistry of these compounds offers perspectives on their actions regarding performance, safety and environment [28]. There are a couple of main ways through which cyanide can damage the environment: leakages, spills and evaporation from open leaching basins [21]. To reduce the negative impact on the environment, the naturally occurring cyanide dissociation mechanisms such as volatilization, complexation, precipitation, adsorption, biological transformation, and sulfidation can be artificially enhanced [27]. By complexation, most often with formation of iron cyanocomplexes, cyanide can be removed from wastes. However, considering that these complexes can dissociate photochemically into free cyanides, some concerns regarding the environmental risks were raised [27]. The adsorption of weak and strong cyanometallic complexes on goethite, manganese oxides, aluminum oxides, silica, and clays is another option for cyanide reduction [27]. Biological transformations assisted by bacteria, fungi or plant species may break down cyanide via oxidative, reductive, hydrolytic or assimilatory pathways to carbon and nitrogen species used for their growth [27]. The rate of cyanide attenuation in tailings is influenced by cyanide speciation, pH of the solution, temperature, redox conditions, exposure to sunlight and microbial activity [27].

2.2 Thiourea leaching

Thiourea (SC(NH₂)₂) dissolves gold by forming a complex in acidic solution according to Eq. (2) [20, 22, 29, 30]. In the leaching process, ferric sulfate is added as a catalyst to enhance the gold oxidation [20, 22].

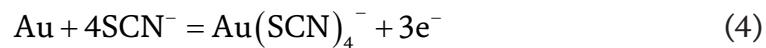
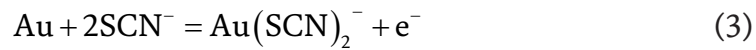


Gold dissolution in thiourea is faster than in cyanide, but high volumes of thiourea solutions are needed in the dissolution process [22]. This shortcoming is avoided by using a mixture of thiourea, thiocyanate and ferric sulfate [21]. Thiourea presents low sensitivity to base metals present in mining wastes and tailings used as raw material for Au recovery [29]. Regardless of its low toxicity compared to cyanide, thiourea is classified as a potential carcinogen [20, 29].

Extensive research has been carried out on Au leaching using acid thiourea solution at laboratory scale. Moreover, thiourea leaching of Au was tested in pilot plants and industrial operations [28, 30]. The pH of the solution, leaching potential, ferric sulfate concentration, thiourea concentration and leaching time are critical parameters for the leaching efficiency [20]. The optimum conditions for thiourea leaching can be achieved by using an appropriate quantity of oxidant to ensure the oxidation of 50% of the thiourea to formamidine disulfide. An excessive amount of oxidant will lead to high thiourea consumptions. High consumptions combined with the use of reagents for pH adjustment and potential control will increase the cost of the leaching process [28].

2.3 Thiocyanate leaching

Thiocyanate (SCN⁻) forms two stable and soluble complexes with gold, Au(SCN)₂⁻ (aurothiocyanate) and Au(SCN)₄⁻ (aurithiocyanate), out of which Au(SCN)₄⁻ is the most stable (Eqs. (3) and (4)) [28, 31].



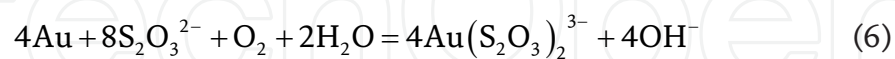
Thiocyanate has low toxicity, high stability, but also slow leaching rate. Thus, ferric ion as an oxidant is used to increase the leaching rate. Gold leaching using thiocyanate takes place in the presence of ferric ions, according to Eq. (5) [19].



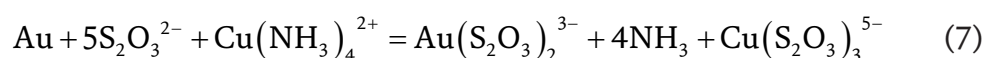
The thiocyanate/ferric sulfate system proved to be suitable for a wide range of ferric ion/thiocyanate molar ratios and slowed the rate of thiocyanate decomposition [32]. Generally, gold leaching using thiocyanate (0.01–0.05 M) occurs at a potential of 0.4–0.45 V, pH 1–3, in the presence of an oxidant like ferric ions (2–5 g/L) or peroxide. It has been shown that, under optimal conditions, a gold extraction yield of 95% can be achieved [21]. Thiocyanate concentrations in the range of 0.5 to 5 g/L and Fe (III) concentrations in the range of 6 to 12 g/L have been used in laboratory and small-scale pilot tests. The thiocyanate concentration must be maintained for the effective leaching of Au [28]. Thiocyanate leaching has not yet been commercialized due to its complex operating process [32]. As the leaching takes place at low pH, special reactors are required to resist in highly corrosive and oxidizing media [29]. Before its large-scale implementation, more research is needed to identify the optimum conditions of the process and to reduce the thiocyanate consumption.

2.4 Thiosulfate leaching

The thiosulphate leaching proved to be a very promising, environmental-friendly alternative to cyanide leaching for Au and Ag recovery [20]. Usually it is used under alkaline conditions to avoid thiosulfate decomposition in the presence of oxygen as oxidation agent (Eq. (6)) [20, 21, 28].



Generally, the dissolution rate of Au in alkaline thiosulphate is slow, but it can be enhanced in the presence of ammonia and Cu^{2+} [20, 21]. The stability of thiosulfate and copper complexes depends on the pH of the solution [22]. The Au dissolution in ammoniacal-copper thiosulfate takes place according to Eq. (7) [28].



The main advantages of thiosulphate leaching compared with other methods are the low toxicity, high reaction selectivity, ability to recirculate the leaching solutions, lower reagent cost and the possibility to recover the dissolved gold by adsorption and electrodeposition [8, 33]. The main disadvantage of alkaline thiosulphate leaching is the high reagent consumption and low extraction rates [21].

Recently, Ubaldini investigated the use of thiosulphate for Au extraction using mine tailings from Ribita and Criscior [8]. A gold extraction of 75% after leaching and a recovery of 90% Au after purification (adsorption–desorption–electrodeposition cycle) was obtained, while the overall process achieved a recovery of 65–67% Au [8]. The Au and Ag recovery from mineral tailings using glycine and sodium thiosulphate showed a leaching recovery of Au more than 80% after 48 h leaching tests with thiosulfate at a solid–liquid ratio of 1:1 [34].

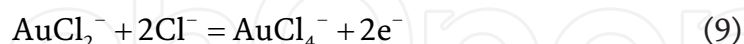
Several studies have been carried out over the last decades to establish the commercial feasibility of Au leaching using thiosulphate. The high consumption of thiosulfate during Au leaching represents a disadvantage for scaling up the process as thiosulphate is easily oxidized by the copper ions added as catalyzer [33]. Moreover, the low affinity of activated carbon for the gold complex makes the recovery of Au more difficult [33].

Mahmoud and Awad studied the recovery of Au from thiosulfate solution on activated carbon in the presence of ammonium persulfate [35]. The obtained results showed that increasing the ammonium persulfate concentrations to 0.01 M, an efficiency of 85% for gold adsorption after only 10 min was obtained, and 95% of Au was recovered after 90 min [36]. The recovery of Au from the thiosulphate leachate using strong base anion resins compared with weak base anion resins presents several advantages such as higher adsorption capacities of gold complex and independence of the adsorption performances on the pH of the solution. However, their selectivity for $[\text{Au}(\text{S}_2\text{O}_3)_2]^{3-}$ against $[\text{Cu}(\text{S}_2\text{O}_3)_3]^{5-}$ is low and complex processes are required for the complete separation of gold and copper from the anion resin [33].

2.5 Halide leaching

2.5.1 Chlorination

Before the large-scale use of cyanide leaching, chlorination was widely applied for gold recovery [20, 21]. Gold dissolution occurs in two stages according to Eqs. (8) and (9) [28].



Compared to alkaline cyanide leaching, chlorination offers a high dissolution rate, but requests acidic media, high temperatures and high concentrations of chloride [21]. In the case of minerals containing silver and lead the metal recovery is low. Moreover, during the leaching process highly toxic and corrosive chlorine gases are released [21]. The reagent consumption is high when low concentrations of sulfides are present in the wastes, leading to the reduction of the gold complex to metallic gold [20].

Recently, Ahtiainen and Lundstrom studied cyanide-free gold leaching in mild chloride media obtaining a gold extraction of 72% at a redox potential <520 mV vs. Ag/AgCl [37].

The combination of chlorination with roasting for the Au extraction showed that 91.6% of Au was recovered using 4% CaCl_2 , while heating at 1323 K for 2 h [38]. Li studied the gold extraction from tailings using sulfuric acid as a pretreatment before calcium hypochlorite leaching [39]. The obtained results showed that the leaching rate of Au was 81% using 8% calcium hypochlorite, 333 K chlorination

temperature and 2 h chlorination time [39]. The use of chlorine for gold extraction remains difficult to use due to the high reagent consumption, its corrosive effect, high cost of the overall process and challenges experienced in Au recovering process [28].

2.5.2 Bromine

Gold dissolution in bromide (Eq. (10)) is influenced by the concentration of bromide and Au, the pH and the electrochemical potential of the anodic and cathodic processes [21, 40].



Bromide leaching has the advantages of short extraction time, high dissolution rates, high selectivity and adaptation to a wide range of pH values. However, bromide is not suitable to be used as a large-scale industrial process as it is difficult to handle and has high reagent costs [20–22, 40].

Over the last years, gold dissolution was investigated using different oxidants like ferric ion, hydrogen peroxide and hypochlorite. The results showed a slow dissolution of gold using hydrogen peroxide and ferric ion [40]. The efficiency of bromine produced in-situ using NaBr, NaOCl and HCl to dissolve auriferous gold ore from the Castromil deposit (Portugal) was proven to be comparable with that of cyanide leaching and twice higher than of thiosulfate [40].

2.5.3 Iodine

Triiodide ion formed by iodine (I₂) and iodide ions (I⁻) acts as an oxidant complexing gold (Eq. (11)) [21, 29].



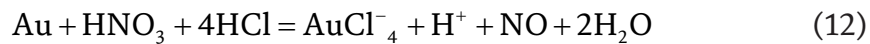
Iodine gold leaching can be a promising alternative to cyanide leaching because of its low volatility and hazardousness [22]. The process presents high leaching rates and the formed gold iodide complexes are more stable in aqueous solutions than the complexes with other halogens [20]. However, iodine was not used at industrial scale due to its high cost [22]. The electrolytic deposition could be an efficient method to reduce the costs through electrolytic recovery of gold and regeneration of iodine [29]. Recently, electrodeposition of gold from iodine leaching solution using response surface methodology was used to study the interactions between variables and their effect on gold deposition rate. The proposed method showed a high deposition rate (94.02%), but a low coulombic efficiency (2.53%) [41].

Presently, the application of halide systems is limited due to the difficulties encountered in maintaining the gold complex in the solution [20].

2.6 Aqua regia leaching

Aqua regia is a mixture of concentrated nitric acid (HNO₃) and hydrochloric acid (HCl). The dissolution of gold with aqua regia is a simple, fast and effective process, but the amount of NO_x released in the atmosphere can be a significant

source of air pollution [42]. The HNO_3 favors the formation of trivalent gold ions, which further react with chloride to form tetrachloroaurate anions, according to Eq. (12) [21, 43].



The aqua regia leaching is mostly used for alloys with high gold content [36]. In case of higher Ag content, it is necessary to reduce the amount of Ag to prevent the incomplete dissolution of gold by blending doré materials with feedstocks containing low amounts of Ag or by pretreatment with HNO_3 for Ag removal before dissolution. Aqua regia is known as an effective leaching agent due to its high dissolution rate, but is extremely corrosive [21]. During the leaching process, toxic NO_x gases are released. The NO_x emissions depend on the acid concentrations, temperature and air flow rates of the dissolution process [42]. The effectiveness of the dissolution process in aqua regia is influenced by the material granulation which can offer a large surface area to enable the reaction kinetics. In addition, the solution can be heated during dissolution to allow a rapid reaction.

2.7 Microbial leaching

Recovery of value-added metals from various wastes using microbial species has attracted much interest in recent years due to the long-term decline of ore grades and concentrates. Microorganisms play an important role especially in the gold recovery process from mining wastes and tailings. Some specialized bacteria, fungi, yeasts, algae or actinomycetes are increasingly being used to facilitate the extraction of gold from low-grade auriferous ore [44]. These microorganisms can enhance the oxidation of metallic minerals and may be used as flotation agents or as biosorbents in the gold recovery process [24, 45].

In recent years, two different types of biomining processes have attracted researchers' interest as alternatives to conventional methods: bioleaching and biological oxidation, as they proved to be cost-efficient, sustainable and non-hazardous [46–48].

Bioleaching is a solubilization process in which bacteria help to dissolve gold from ores or wastes, while in the biooxidation process, the acidophilic microorganisms release gold from minerals during sulfide oxidation [49]. Biooxidation of gold can be applied as heap or dump leaching and stirred tank leaching. The static biooxidation techniques are based on the principle of circulating water and air through waste heaps to activate the growth of microorganisms that amplify the oxidation [50]. Cyanogenic microorganisms as *Chromobacterium violaceum*, *Pseudomonas fluorescens* and *Pseudomonas plecoglossicida* were able to mobilize gold when grown in the presence of various metal-containing solids. Compared to chemical oxidation, biological oxidation offers the advantages of low production costs, low temperatures, low pressures, partial sulfide oxidation, decreased leachate consumption and no atmospheric pollution [51]. In polymetallic sulfidic ores several acidophilic, chemolithotrophic iron and sulfur oxidizing bacteria are present. Mesophilic iron and sulfur oxidizing bacteria as *Acidithiobacillus ferrooxidans*, sulfur-oxidizing *Acidithiobacillus thiooxidans*, iron-oxidizing *Leptospirillum ferriphilum* and *Leptospirillum ferrooxidans*, moderately thermophilic bacteria, such as sulfur-oxidizing *Acidithiobacillus caldus* and sulfur and iron oxidizing *Sulfobacillus* spp. has been reported to assist in the oxidation of sulfides [44, 52].

Various microorganisms were tested for gold biomining [51]. A recent study has reported the Au recovery from polymetallic sulfide minerals using biooxidation followed by acid washing and citrate leaching [53]. Thus, the biooxidation stage removed the refractory ores, increasing the gold extraction from 17.3 to 86.4% [53]. In the biooxidation process, sulfides are oxidized by mixed mesophilic culture mainly consisting of *Acidithiobacillus ferrooxidans*. A combined procedure for Au, Ag and Pb recovery from sulfide minerals was proposed by Lorenzo-Tallafigo [54].

Biosorption by microbial biomass is another promising, cost efficient and eco-friendly method for the gold recovery from wastes [23, 54]. Biosorption is a passive sorption and/or complexation method of gold in the cell wall of diverse algae, fungi and bacteria biomass [54, 55]. Stationary or dead microbial biomass bind and concentrate gold ions from pregnant leachates [56]. In biosorption processes a series of green and brown algae (*Chlorella vulgaris*, *Fucus vesiculosus*, *Sargassum natans*), fungi (*Aspergillus niger*, *Mucor rouxii*, *Rhizopus arrizus*, *Aspergillus oryzae*, *Chaetomium globosum*, *Gibberella fujikuroi*, *Mucor hiemalis*, *Penicillium chrysogenum*, *Purpureocillium lilacinum*), yeast (*Candida krusei*, *Candida robusta*, *Candida utilis*, *Cryptococcus albidus*, *Cryptococcus laurentii*, *Debaryomyces hansenii*, *Endomycopsis fibuligera*, *Hansenula anomala*, *Hansenula saturnas*, *Kluyveromyces* spp., *Saccharomyces cerevisiae*, *Sporobolomyces salmonicolor*, *Torulopsisaeria*) or bacteria (*Streptomyces phaeochromogenes* HUT6013, *Acinetobacter calcoaceticus*, *Erwinia herbicola*, *Pseudomonas aeruginosa*, *Pseudomonas maltophilia*) are used for gold recovery [24]. Some microbial species (*Bacillus subtilis*, *Escherichia coli*, *Streptomyces albus*, *Candida utilis*, *Aspergillus niger*) can contribute to the passive sorption of gold from solution or have the capacity to accumulate gold in an EPS capsule (*Hyphomonas adhaerens* MHS-3) [51]. A gold recovery rate of 85% has been reported by Kenney when non-metabolizing bacteria cells of *Bacillus subtilis* and *Pseudomonas putida* were used [57].

Recent studies revealed effective results for Au recovery with other biomining methods as bioprecipitation, biomineralization, bioflotation, bioflocculation, biosorption and bioaccumulation [58–60]. All these methods are based on gold harvesting microorganisms, which are either isolated from the gold enriched areas and domesticated or bioengineered strains with exceptional gold retrieval efficiency [48].

Due to the fact that various microbial communities have different structure, functions and dynamics in the gold metabolism, their use in bio-hydrometallurgical processes still remain a challenge. Even if many thermophilic or acidophilic bacteria, archaea or other types of microorganisms have been isolated, characterized, and even used for extracting precious metals, new strains isolated from different sources of wastes are needed [47]. At the same time, it is necessary to find combinations of chemicals compatible with efficient microbial agents to recover high levels of gold [61].

In the mining industry, naturally-occurring microorganisms which can be exploited through different strategies for the extraction and recovery of gold have a great potential [62]. The use of microorganisms for the recovery of precious metals from waste is economical and can avoid environmental pollution [44]. Furthermore, microbe driven technologies based on processes like biofiltration using specialized biofilters can assure a specific recovery of the gold ions [63]. Future gold recovery processing systems based on microorganisms will revolutionize the gold production.

3. Conclusions

Mining wastes represent a valuable resource for various elements, among which precious metals like gold. These wastes present both challenges and opportunities,

as it require long term management to reduce the environmental risks, but could act as important resources for base and precious metals. In order to secure the high global demand for gold, new, low-cost, highly efficient but also environmentally friendly methods need to be developed. Although there is a lot of research in the field of developing less-toxic alternatives to cyanide and several leaching agents proved their efficiency, till now, there is no viable industrial scale alternative to cyanide. Another important challenge is the recovery of Au from mine waste leachates containing several soluble metals in various concentrations.

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Conflict of interest

The authors declare no conflict of interest.

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