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Highly efficient and selective leaching of silver from electronic scrap in the base-activated persulfate – ammonia system

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ABSTRACT

A system composed of persulfate salt and ammonia in highly alkaline aqueous solution is developed and examined for leaching metallic silver from elements of the electronic waste materials (e-scrap). Strong base activates persulfate ions providing in situ generation of highly reactive oxygen molecules. The oxidized metal forms then well soluble complex ions with ammonia ligands. The kinetic studies of the leaching process were performed for pure metallic silver. They revealed that the efficiency of the process is affected by the type of the persulfate salt. By employing potassium persulfate one obtains significantly (more than 50% for silver plates and more than 100% for silver powder) increased efficiency of silver dissolution compared to the solution composed of either sodium or ammonium persulfates. In the range of persulfate concentrations between 0.02 and 0.23 mol/L the apparent reaction order with respect to the persulfate concentration was similar for all persulfate salts and was estimated to be around 0.5. The room temperature (22 ± 2 °C) seems to be an optimal temperature for the leaching process. An increase in the temperature resulted in the significant drop in the silver dissolution rate due to the decreased solubility of oxygen. Based on these results a possible mechanism of dissolving silver is discussed and the optimal composition of the leaching solution is formulated. The obtained formulation of the leaching solution was applied for the extraction of silver coatings of Cu-based e-waste scrap and the obtained results revealed an important effect of copper in the mechanism of the leaching process. The regression analysis of the leaching curve indicated that each gram of base-activated potassium persulfate under the specified conditions may leach almost 100 mg of silver coatings in a form of well soluble diamminesilver (I) complex. The silver complex can be relatively easily reduced to metallic silver. The method developed is relatively cheap, low toxic and does not produce harmful by-products.

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1. Introduction

Large amounts of electronic scrap usually called “e-waste” are actually a very serious threat to the natural environment. Electronic scrap is generated by discarding of used electrical and electronic equipment such as the computer hardware, printed circuit boards, connectors and electric devices. Scrap from these materials is usually characterized by a thin layer of a single precious metal (mainly Ag, Au, Pd) deposited or plated on a substrate usually made of copper, nickel, aluminum or steel. The recovery of precious metals is the main economic driving force for the recycling of e-waste materials. Of these metals gold and silver receive the most attention. The former due to its high price and considerable usage in the electronic industry and the latter – because of its abundant and common presence in various types of electronic ele-

ments. The treatment of electronic scrap usually proceeds in two steps. In the first step the scrap material is roughly categorized and involves removal of bulk material. In the second stage various metals are concentrated and then recovered. The recovery of precious metals from electronic scrap may involve various processes (Cui and Zhang, 2008). These include pyrometallurgy, biotechnology, electrochemistry and hydrometallurgy or extractive metallurgy (Tuncuk et al., 2012).

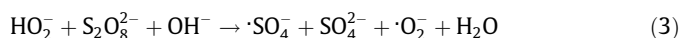
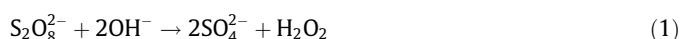
There are many chemical systems that are capable to leach efficiently both: gold and silver. The most efficient system is the one composed of cyanide solutions and oxygen as oxidant (Marsden and House, 1992). Considerable toxicity of cyanide solutions to humans and environment triggered an effort for seeking alternative, less harmful and aggressive, systems to leach these metals (Alymore and Muir, 2001). Among the most promising are copper (II) – ammonia – thiosulfate (Briones and Lapidus, 1998; Jared and David, 2013; Ibarra-Galvan et al., 2014), copper (II) – ammonia – thiosulfate – EDTA (Puente-Siller et al., 2013), iron (III) – thiourea

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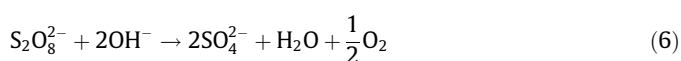
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(Li et al., 2012), ultrasound assisted sulfuric acid – thiourea (Salim et al., 2005), oxygen (ozone) – thiosulfate (Viñals et al., 2005; Rodríguez-Rodríguez et al., 2014; Rivera et al., 2015), nitric acid (Holloway et al., 2004), and halide (Almeida and Amarante, 1995; Shaohua et al., 2011; Zhang et al., 2012) systems. In these systems the role of the oxidant for the noble metal plays either Cu^{2+} or Fe^{3+} cations or strong acid (nitric, sulfuric) or oxygen under the appropriate pressure while either thiosulfate or ammonia or thiourea or chloride anions are ligands that form a stable complex with the oxidized metal. These systems are definitely less toxic than the system based on cyanide ions. However, the major concern is related to the stability of the sulfur-based ligands (thiosulfate, thiourea) in aqueous solution. Thiosulfate anion readily decomposes in aqueous solution. Moreover, it is relatively easy oxidized by copper (II) and/or oxygen during leaching to tetrathionate ($\text{S}_4\text{O}_6^{2-}$), trithionate ($\text{S}_3\text{O}_6^{2-}$), sulfate (SO_4^{2-}) and other polythionates. These reactions cause the consumption of thiosulfate anions. To reduce these disadvantageous features the reaction conditions (temperature, pH, concentration of the oxidants) must be constantly controlled and maintained at appropriate levels. In addition to this the chemistry of the thiosulfate – ammonia – copper(II) ion system is complicated due the simultaneous presence of the $\text{Cu}(\text{NH}_3)_4^{2+}/\text{Cu}(\text{NH}_3)_2^+$ redox couple and thiosulfate complexes of silver and copper. A critical comparison of the existing methodologies for the leaching and recovery of silver from different sources has recently been presented in a review article (Syed, 2016).

In this work a new system composed of the persulfate salt and ammonia in highly alkaline aqueous solution is developed and examined for leaching metallic silver. The system employs activated persulfate. The activated persulfate is commonly used in field applications for treating subsurface organic contamination of the soil (Hulling and Pivetz, 2006). Persulfate acts as an oxidizing agent providing in situ non-specific destructive oxidation of most organic contaminants (Watts and Teel, 2006). The strong oxidation properties of persulfate ions can be activated using either chelated iron (Block et al., 2004), iron nanoparticles (Zhu et al., 2016) copper ions (Liu et al., 2012; Liang et al., 2013) or a strong base (Furman et al., 2010). The iron and copper activation methods promote the decomposition of persulfate species to highly reactive sulfate radicals while the method involving the strong base leads directly to the generation of reactive oxygen species. The mechanism proposed for the base-activated persulfate decomposition may be represented by the following set of reactions (Furman et al., 2010):



The set of Eqs. (1)–(5) can be summarized by the following net equation:



The proposed mechanism implies strong dependence of the effectiveness of the oxidation processes involving persulfate on pH. In the absence of competing activating species (e.g. metal cations) the reactivity of persulfate ions may decrease dramatically at $\text{pH} < 10$ (Block et al., 2004; Furman et al., 2011).

In recent years some strategies for the recovery of gold from gold-coated elements of waste electrical and electronic equipment

using persulfate salts have been proposed (Syed, 2006; Alzate et al., 2016). In these approaches the role of persulfate was to oxidize and leach the metallic substrate (Ni, Fe, Cu), leaving gold coatings in a form of solid flakes. The oxidation process was strengthened by increasing temperature and addition of oxygen at the specified flow rates.

The objective of this work was at first to take advantage of high reactivity of the base-activated persulfate system for the selective oxidation of metallic silver layers deposited on copper base elements. The proper understanding of the process and the optimization of the composition of the leaching solution required detailed kinetic studies of the process of metallic silver dissolution in the base-activated ammonia solutions of the persulfate system. The effects of persulfate concentration, type of persulfate salt, and temperature were investigated. Based on these results a possible mechanism of dissolving silver is discussed. Finally, the optimal composition of the leaching solution that minimizes reagent consumption and maximizes the efficiency of silver extraction is formulated.

The procedure was examined for leaching silver layers deposited on copper-based substrates which represent real e-scrap samples. The method was tested using only one type of e-waste materials presented in Fig. 1 A. These e-waste elements are commonly used as grounding components in many electrical devices. They were collected from waste radio station equipment.

2. Materials and experimental procedure

2.1. Materials

All chemical constituents of a leaching solution (sodium hydroxide (NaOH), 25% aqueous solution of ammonia (NH_3), ammonium persulfate ($(\text{NH}_4)_2\text{S}_2\text{O}_8$, APS), sodium persulfate ($\text{Na}_2\text{S}_2\text{O}_8$, SPS), and potassium persulfate ($\text{K}_2\text{S}_2\text{O}_8$, PPS), POCh, Gliwice, Poland) were of at least 99% purity. They were used as received. Silver plates (99.9%) with 1 mm thickness and a 1.5 cm^2 surface area were used in kinetic experiments while a silver powder (99.9%, Sigma Aldrich) consisting of spheroidal grains (of approximately 1 μm in diameter) was used for supporting experiments. In the application experiments copper plates (from waste radio station equipment) covered with a silver galvanic layer were employed. Two representative samples are shown in Fig. 1A. The e-waste employed for the studies was chemically and physically homogeneous material. It was composed of copper covered with a thin layer of silver. The dimensions of the plates were: 0.5 $\text{cm} \times 3 \text{ cm}$. The mean content of silver was less than 3% per element. Silver was present in the form of a layer of a 2 μm thickness. The average weight of one element was 0.35 g. Silver nitrate (AgNO_3 , Sigma Aldrich, 99.9%) was used for the construction of a calibration curve for X-ray fluorescence spectrometry (XRF). Ultra pure water (Milli-Q system, Millipore) was used in all experiments.

2.2. Experimental procedure

The leaching system consisted of an open 250-mL round-bottom flask (reactor) with magnetic stirring. The reactor was kept in the constant temperature in a water bath under atmospheric pressure. The temperature of the water bath was controlled using a refrigerated circulator (Polystat, Cole Parmer). The sample of metallic silver either in a form of a plate or powder or a layer deposited on the copper plate was placed in the leaching solution. The latter solution was initially composed of 0.5 mol/L NaOH and 1.2 mol/L NH_3 dissolved in 110 mL of aqueous solution. The temperature was adjusted to the desired level (either 22 $^\circ\text{C}$ or 8 $^\circ\text{C}$ or 38 $^\circ\text{C}$ or 53 $^\circ\text{C}$). Finally, a known amount of a solid persulfate salt

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