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Recovery of gold from waste electrical and electronic equipment (WEEE) using ammonium persulfate

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ABSTRACT

This paper presents a novel methodology to recover gold from waste electrical and electronic equipment (WEEE) using ammonium persulfate ($(NH_4)_2S_2O_8$). Gold was recovered as a fine coating using substrate oxidation without shredding or grinding process. The WEEE sample was characterized giving values of Au: 1.05 g/kg, Fe: 86.00 g/kg, Ni: 73.64 g/kg, Cu: 26.65 g/kg. The effect of $(NH_4)_2S_2O_8$ concentration (0.22–1.10 M), oxygen (0.0–1.4 L/min) and L/S ratio (10–30 mL/g) on the main responses (substrate oxidation and Au recovery) was investigated implementing response surface methodology with numerical optimization. A quadratic model was developed and quantities greater than 98% of Au were recovered. The findings presented suggest that, optimized quantities of ammonium persulfate in aqueous highly oxygenated media could be used to extract superficial gold from WEEE.

1. Introduction

Non polluting methodologies of valuable metals recovery from secondary sources have been a current topic in several studies because of their low environmental impact. Many of these studies have focused on recovering gold avoiding the hazardous implications of mineral extraction such as water pollution, deforestation and health repercussions. Nowadays, one of the main secondary sources to recover precious (Au, Ag, Pd) and base metals (Cu, Ni, Fe) is waste electrical and electronic equipment (WEEE) (Yazici and Deveci, 2014). The rapid growth in the manufacturing of technological devices has generated large quantities of WEEE. The annual WEEE global growth has been estimated at 8.8% (2004–2011) and 17.6% (2011–2016) calculating for 2016 a global volume of 93.5 millions of tons (Akcil et al., 2015; Yu et al., 2014). These values expose the adequate final disposal of WEEE as one of the main challenges in WEEE management due to waste of valuable metals (Hadi et al., 2013) and the environmental problems associated with conventional disposal and recycling methods.

Conventional disposal methods of WEEE such as landfill, incineration and hydrometallurgical recycling techniques generates soil and water pollutants and harmful substances that are released into the air. On the first hand, landfill and incineration produce toxic substances that comprehend Hg, Pd, Cd, dioxins, furans and heavy metals vapors (Yazici and Deveci, 2013). However, developing countries still use landfill and incineration causing public health and environmental risks. Risks of landfill leachate production and air, soil, water or underwater pollution, make these disposal process unsuitable (Hadi et al., 2015a). On the other hand, hydrometallurgical recycling techniques with strong acids (HCl, HNO_3, H_2SO_4/H_2O_2) or oxidative reagents (cyanide, thiourea, halide, nitrate and iodide) have been suggested by a variety of authors to process WEEE and reach the recovery of gold (Birloaga et al., 2014; Petter et al., 2014; Shibayama et al., 2013). Methods of gold recovery from WEEE with hydrometallurgical techniques focus on primary physical separation (shredding, grinding) and secondary leaching of total metallic fraction (Bas et al., 2014). The metallic fraction (precious and base metals) is obtained after shredding and grinding, which are recognized by dust pollution generation (Naseri Joda and Rashchi, 2012). In addition, grinding fraction is dissolved using strong acids or oxidative reagents (Zhang et al., 2012) and the resulting solution is separated through time-consuming chemical process (cementation, solvent extraction, precipitation or coagulation) with the aim to recover the metal of interest from the solution (Syed, 2012). Hydrometallurgy extensively use cyanide, thiourea, halides and some strong acids. These agents are recognized by its toxic potential, low chemical stability and environmental impact (Tuncuk et al., 2012).

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Nowadays, strategies on WEEE management have been adopted to mitigate health and environmental problems. The strategies contemplate the development of tools as Material Flow Analysis (MFA), Multi Criteria Analysis (MCA) and Life Cycle Assessment (LCA) (Kiddee et al., 2013). In develop countries, MFA and MCA have been successful applied to estimate the generation of WEEE and take environmental decisions to solve multi-criteria problems on disposal, while LCA has been used in several studies to evaluate the environmental impacts of WEEE (Wäger et al., 2011). Studies on LCA concluded that, compare with landfill or incineration, recycling techniques are more appropriate to manage WEEE (Kiddee et al., 2013). Nevertheless, any recycling process perform without environmental care may produce a highest impact on soil, air, water and humans. Developing suitable and optimal recycling methodologies capable of avoiding pollution, reducing reagents toxicity and time-consuming reactions has become one of the most important topics in WEEE management research.

Hence, in recent years, some methodologies that attempt to use environmentally friendly agents and the optimization of gold and base metals recovery from WEEE have been suggested (Barbieri et al., 2010; Ha et al., 2014; Syed, 2006). For instance, alternative agents to extract non-leaching gold that include potassium persulfate ($K_2S_2O_8$) (Syed, 2006) and cupric chloride ($CuCl_2$) (Barbieri et al., 2010) were studied to recover gold from WEEE. These agents were used to oxidize and leach the metal substrate (Ni, Fe, and Cu) where gold was superficially associated as coating (Barbieri et al., 2010). The partial leaching of the substrate permitted gold recovery in a solid particulate state (Barbieri et al., 2010; Syed, 2006). Due to the elimination of gold leaching, it was possible to reduce reaction time avoiding purification stages and achieving the 98% in Au recovery with minimum formation of contaminant by-products or total agent regeneration (Barbieri et al., 2010; Syed, 2006). Redox potential for the production of (SO_4^{2-}) ions in aqueous potassium persulfate was estimated in 2.01 V (Huang et al., 2002) and 0.48 V for the production of $(CuCl_2)^-$ ions in acid solution (Lundström et al., 2009). These estimations exposed the ability of $K_2S_2O_8$ and $CuCl_2$ to oxidize Ni, Fe and Cu and release gold from the substrate. Despites its great advantages, persulfate and cupric chloride oxidative systems to recover non-leaching gold from WEEE have not been extensively investigated.

Selective persulfate oxidation proposed by Syed (2006) catalyzed with oxygen can be optimized in order to maximize the recovery of gold from WEEE reducing parameters like agent consumption and reaction time (Birloaga et al., 2013; Hadi et al., 2015b; Jordão et al., 2016). A way to optimize this process is to use response surface methodology (RSM), a series of statistical techniques. RSM has been adopted in several studies to find optimal region of operation into an experimental design space (Montgomery, 2013). This methodology was reported in WEEE management to determine the greater amount of gold leaching evaluating the incidence of thiosulfate, cooper and ammonia concentration (Ha et al., 2014). Besides, the extraction of Cu, Fe, Ni, Ag and Pd from waste printed circuit boards (WPCBs) was studied adopting RSM to establish the conditions that maximize metals extraction using H_2SO_4–$CuSO_4$–NaCl solutions (Yazici and Deveci, 2013).

In this study, an environmentally friendly methodology to extract non-leaching gold by partial substrate oxidation from WEEE using ammonium persulfate $(NH_4)_2S_2O_8$ was developed. The incidence of $(NH_4)_2S_2O_8$ concentration, oxygen and liquid/solid ratio on the recovery of gold was analyzed using response surface methodology with numerical optimization. The effects of $(NH_4)_2S_2O_8$ concentration (0.22–1.10 M), oxygen (0.0–1.4 L/min) and liquid/solid ratio (10–30 mL/g) over the gold recovery were studied in five different levels through a central composite design (CCD). $(NH_4)_2S_2O_8$ was selected over other environmentally friendly agents due to the lack of research on its use and optimization in recovering gold from WEEE through substrate oxidation. In addition, the produced persulfate ions ($S_2O_8^{2-}$) are not absorbed or bio accumulated in the soil after the process (Hernandez, 2005) and the generated by-products (sulfates) have not a negative effect on the environment (Syed, 2006). Comparing with potassium persulfate, sodium persulfate and cupric chloride, $(NH_4)_2S_2O_8$ has a greater leaching power to oxidize base metals than potassium and sodium persulfate (Babu et al., 2002) and is operationally safer and less toxic than $CuCl_2$. The toxicity reduction responds to the absence of Cl_2 (g) during the reaction.

In brief, the system of methods applied was carried out without shredding or grinding stages which reduced secondary dust pollution, reaction time and contaminant by-products. This study aimed at maximizing gold recovery from WEEE with a novel methodology that includes the partial oxidation of metal substrate with ammonium persulfate in oxygenated media and the numerical optimization of the most significant parameters.

2. Experimental

2.1. Materials and reagents

Intel Celeron–Pentium electronic processor scrap from end-of-life computers supplied by a local recycling company was the sample used (Fig. 1). A total amount of 50 processors were selected by reference, shape, weight, superficial distribution of gold and manufacturer to ensure sample homogeneity and statistical significance. Intel Celeron and Intel Pentium processors of 5×5 cm^2 and an average weight of 8.97 g (Fig. 1) were used for chemical characterization, substrate oxidation and gold recovery tests without shredding or grinding stages.

A sample of 35 g was used to determine the amount of gold and metal substrate (Fe, Ni, Cu) by chemical digestion using aqua-regia (Lee et al., 2011; Petter et al., 2014) followed by microwave plasma – atomic emission spectroscopy (MP-AES, AGILENT 4100) (Table 1). After chemical characterization, aqueous commercial grade ammonium persulfate ($\geqslant$98% $(NH_4)_2S_2O_8$) with a water solubility of 850 g/L at 25 °C (Hernandez, 2005) was the selected environmental reagent used to produce sulfate ions (SO_4^-), which partially oxidized the metal substrate breaking the Au–Cu–Ni–Fe bond and allowing gold to be extracted in its original non-leaching state.

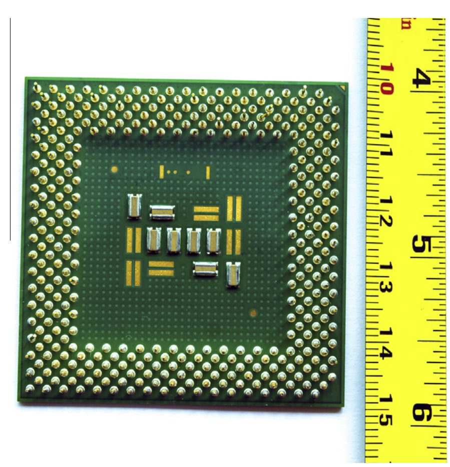

Fig. 1. Processor scrap sample (ref. Intel Celeron–Intel Pentium).

An oxygen tank (99.9% O_2) with flow control was used to deliver O_2 and catalyze the oxidative reactions.