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## Initial studies about gold leaching from printed circuit boards (PCB's) of waste cell phones

Sebastián Camelino<sup>a\*</sup>, Javier Rao<sup>a</sup>, Roger López Padilla<sup>a</sup>, Roberto Lucci<sup>a</sup>

<sup>a</sup> *Department of Metallurgical Engineering, National Technological University, Regional Faculty Córdoba, Maestro M. López esq. Cruz Roja  
Argentina, Ciudad Universitaria, Córdoba Capital, 5000, Argentina.*

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### Abstract

Excessive consumption of mobile phones, a decrease in their life time and, therefore, an increase in the generation of this type of electronic scrap, is noted in recent years. Printed circuit boards (PCB's) from waste cell phones are attractive resources because of the valuable metals Cu, Au, Ag, Pd they possess, but at the same time dangerous due to their content of heavy metals. The treatment of these boards is desirable from an economic and environmental perspective.

In this paper we inform the results achieved on laboratory leaching of gold contained in PCB's from cell phones. The reagents used were ammonium thiosulfate in basic medium and thiourea in sulfuric acid solution. Prior to gold leaching, the plates were subjected to grinding to reduce their size to less than 2 mm and to an oxidizing leaching in sulfuric acid solution to separate the copper. The conditions studied in the leaching test were: thiosulfate molar concentration between 0.08 M and 0.12 M and ammonium hydroxide from 0.1 M to 0.2 M, temperature of 20° C, pH = 10.5 and 15 mM of cupric ion. In thiourea leaching, gold dissolution was studied by varying the concentration of the reagent between 20 g/l to 28 g/l. The results indicate that, under the conditions studied, ammonium thiosulfate is a more effective reagent for the dissolution of gold than thiourea. The gold dissolution values achieved were up to 70% for thiosulfate and no greater than 40% for thiourea.

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\* Corresponding author. Tel.: +54-0351-152563804  
E-mail address: [sebascamelino@gmail.com](mailto:sebascamelino@gmail.com)

## 1. Introduction

Globally, in the last years, it is noticed an increasing consumption of cell phones while their lifetime period is decreasing. This is due to an upward trend in the production of new devices with better functions. Also, there is a trend toward having only a cellphone as a replacement for a landline. For example, in Argentina, almost 10 million of cell phones were discarded since 2008 to 2011, representing 30% of the 34 million phone lines in use. The average lifetime of these appliances is over 2 years, Greenpeace (2011). It is estimated that the number of cell phones will increase in the next years.

Printed circuit boards (PCB's) from discarded cell phones are secondary sources due to the precious and valuable metals included in them. Some of these mentioned metals are Cu, Au, Ag, Pd. Also there are some metals that are dangerous for the environment such as chrome, nickel, cadmium and lead. The treatment of this waste is important since both an environmental and economic standpoint.

Pyrometallurgical processing is the most common way to deal with electronic waste in order to recover copper and other precious metals, Das et al (2009), Cui et al (2008). Because the troubles associated with gas pollution, hydrometallurgical methods are studied. Many reagents have been proposed for leaching of gold, silver and precious metals embedded in e-waste, Huang et al (2009). Cyanide is thoroughly used in gold dissolution from ore. However, its highly toxicity is the main reason to start looking for another alternative. Precious metals dissolution using ammonium thiosulfate and thiourea was investigated in recent years, Tripathi et al (2012), Ficeriova et al (2011), Gurung et al (2012), Ha et al(2010). These reagents represent a new and ecological way for the gold leaching from electronic waste.

In this paper we inform the results achieved on laboratory leaching of gold contained in PCB's from cell phones. The reagents used were ammonium thiosulfate in basic medium and thiourea in sulfuric acid solution. Prior to gold leaching, the plates were subjected to grinding to reduce their size to less than 2 mm and to an oxidizing leaching in sulfuric acid solution to separate the copper.

## 2. Experimental procedure

PCB's from cell phones discarded were used in the leaching experiences included in this work. 120 cell phones have been disassembled with different screwdrivers and pliers. Plastic parts, steel pieces and batteries were disjoined from PCB's as is shown in Fig. 1.



Figure 1 Disassembled cell phone

Fig. 2 shows dismantled boards; they were crashed in order to allow a better surface contact between the reagent

and gold surface, and improving the kinetic of the dissolution. Milling operation was carried on with a cutting mill Retsch, Fig. 3 shows the obtained powder after this step.



Figure 2 PCB's from cell phones

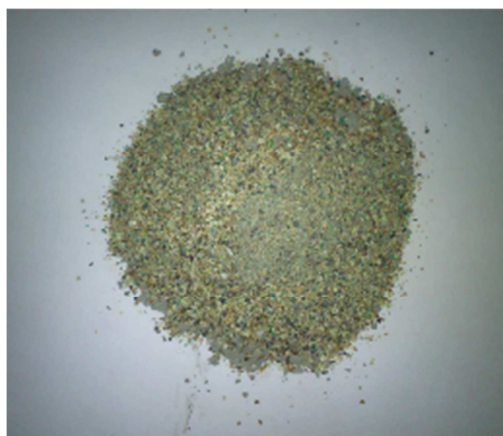


Figure 3 PCB's after milling operation

Two representative samples powder were taken from the whole amount (1200 g) in order to determinate the chemical composition. Each sample had 40 g of weight. Samples were submersed in aqua regia during an hour and then analyzed by ICP-MS. Table 1 shows the reached results after this analysis.

Table 1 Weight composition of metals in PCB's from cell phones

Elements weight %									
Au	Ag	Cu	Ni	Fe	Pb	Sn	Zn	Pd	Pt
0,0168	0,0285	65,74	1,98	1,51	1,065	5,22	0,22	0,011	<0,001

The high content of copper present in PCB's has a negative effect in gold dissolution. In order to avoid the possible interferences due to the presence of copper, it is strongly necessary to dissolve this metal before the gold leaching. Copper leaching was carried on twice with an acid solution of  $H_2SO_4$  catalyzed with  $H_2O_2$ . After first leaching, there was 15,6 % residual copper, which was completely eliminated after second attack.

Leaching experiences to dissolve the gold were carried out with beakers using 5 g of sample with 100 ml of solutions. All experiences were at room temperature ( $\pm 20^\circ \text{C}$ ) with an agitation of 180 rpm. Solutions were filtered after leaching and solid residues were attacked with aqua regia. In all cases, gold dissolution rate was carried on by ICP-MS since the obtained solution of aqua regia and solid residues.

The reagents employed were ammonium thiosulfate, ammonium hydroxide, thiourea, ferric sulfate, cupric sulfate, hydrogen peroxide and sulfuric acid, all of them pro-analysis quality.

### 3. Theoretical aspects

#### 3.1. Dissolution with ammonium thiosulfate

Figure 4 shows the electrochemical-catalytic mechanism for gold leaching in ammonium thiosulfate solutions, Aylmore et al (2001).

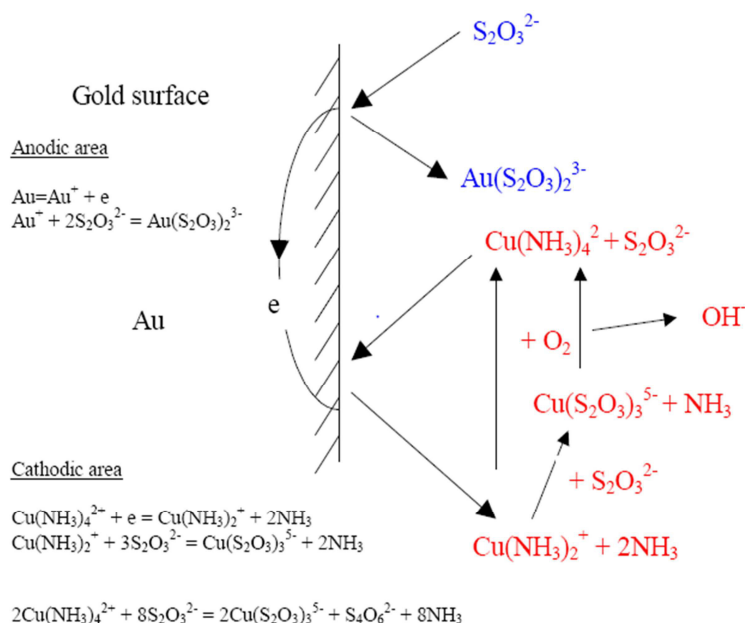


Figure 4 Electrochemical-catalytic mechanism for gold leaching in ammonium thiosulfate solutions

According to this author, gold dissolution is an electrolytic process catalyzed with cupric ions. Gold is dissolved as aurous thiosulfate  $[\text{Au}(\text{S}_2\text{O}_3)_2]^{3-}$  on the anodic area. On the cathodic area, tetra-ammin copper (II) is reduced to  $\text{Cu}(\text{NH}_3)_2^+$  and then, it is re-oxidized to  $\text{Cu}(\text{NH}_3)_4^{2+}$ . Both ammonia and cupric ions are regenerated in this system. Reaction 1 is the result of anodic and cathodic reactions.



Moreover previous reaction, secondary reactions can be produced. These ones could be produced by cupric ions, for example, there is some degradation of thiosulfate ions to tetrathionate as is shown in Reaction 2, Aylmore et al (2001).



In order to avoid reduction of cupric ion to cuprous state and reach an efficient gold dissolution, it is important to keep the correct ammonium and thiosulfate concentrations.

### 3.2. Dissolution with thiourea

Gold dissolution employing thiourea acid solutions with ferric ions as oxidant is shown in Reaction 3, Gonen et al (2007).



Gold is dissolved forming a complex ion with thiourea while ferric ions are reduced to ferrous state. Also, thiourea is easily oxidized to formamidine-disulfure as is shown in Reaction 4.



Formamidine-disulfure is not stable in acid solutions and then it is degraded cyanamide and elemental sulfur according with Reaction 5.



The oxidants amount must be carefully regulated to avoid an excessive thiourea oxidation and, consequently, high reagent consumption is avoided too.

## 4. Results and discussion

### 4.1. Ammonium thiosulfate leaching

The experiences were performed modifying molar concentration of ammonium thiosulfate between 0,07 M and 0,19 M holding the rest of the conditions at the values illustrated in Table 2.

Table 2 Parameters used in ammonium thiosulfate leaching experiences

Parameter				
$[(S_2O_3)^{2-}]$ (M)	$[Cu^{2+}]$ (mM)	pH	Temperature ( $^{\circ}C$ )	Time (h)
0,07 – 0,19	20	10	20	2

Figure 5 shows results obtained with two kinds of samples:

- Crashed PCB's without copper after a double sulfuric acid leaching (curve in blue).
- Crashed PCB's with 15,6 % of residual copper after a single sulfuric acid leaching (curve in red).

As is shown in Fig. 5, gold dissolution rate increases with the raising of ammonium thiosulfate concentration until 0,13 M and then, the percentages of dissolved gold have a slow decrease for greater ammonium thiosulfate concentrations. The same behavior is observed in both curves. Furthermore, samples with copper have lower rates of dissolution than the other ones without the mentioned element. Excessive cupric ion concentration could decrease thiosulfate ionic quantities as a consequence to its degradation to tetrathionate, as was shown in Reaction 2.

It was achieved a good gold dissolution rate, up to 70,5 % in two hours. Despite of that, still is necessary to study the effect of varying other parameters such as: cupric ion concentration, pH, temperature, leaching time and solid-liquid ratio. It is estimated that gold dissolution could be better with the proper conditions in those variables. This will be studied in another stage of researching on the future.

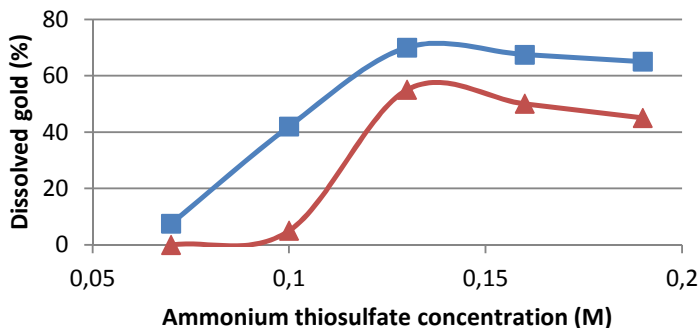


Figure 5 Dissolved gold (%) vs. Ammonium thiosulfate concentration [M]. (pH 10; temperature= 20° C; [Cu<sup>2+</sup>]= 20 mM; leaching time= 2 h.). a) Crashed PCB's without copper after a double sulfuric acid leaching (blue curve) and; b)Crashed PCB's with 15,6 % of residual copper after a single sulfuric acid leaching (red curve).

#### 4.2. Thiourea leaching

There were performed preliminary experiences studying thiourea concentration, with the next set conditions: concentration of ferric ions in 0,6 %, pH = 1 and a period time leaching of two hours. Fig. 6 shows that an increase since 20 g/l to 24 g/l in thiourea concentration raises gold dissolution reaching a top percentage of 37,9 %. To greater values of thiourea concentration than 24 g/l, the dissolution rate decreases. The drop observed in Fig. 6 could be a consequence of a passivation phenomenon generated by elemental sulfur that comes from an excessive thiourea concentration; this mechanism was described in Reaction 5.

Also was studied in another experiences the effect of the leaching time. The thiourea concentration in those experiences was set in 24 g/l. The results are shown in Fig. 7.

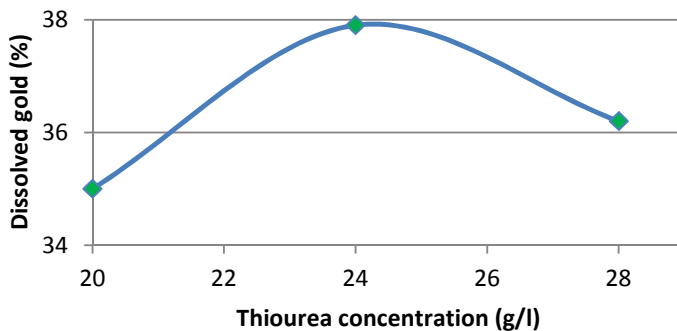


Figure 6 Dissolved gold (%) vs. Thiourea concentration (g/l). [Fe<sup>+3</sup>]= 0,6 %; pH= 1; temperature= 20° C; leaching time= 2 h.

According with Figure 7, the percentage of dissolved gold is 26,8 % for a leaching time of an hour increases to 37,9 % for two hours and with three hours of leaching can be reached 39 % of gold in solution. However, the results already reported are lower than expected. In order to improve these percentages, it is necessary to study the effect of varying other parameters such as: ferric ion concentration, pH and temperature.

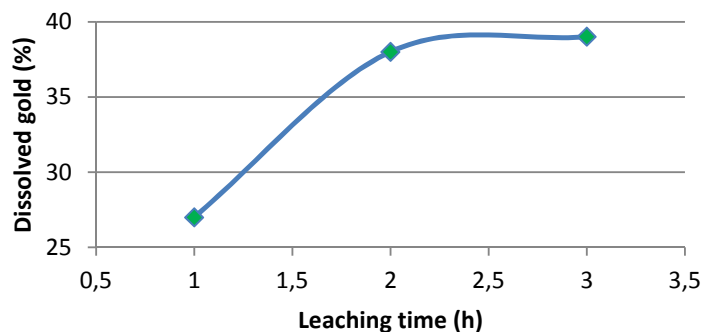


Figure 7 Dissolved gold (%) vs. Leaching time (h). Thiourea concentration= 24 g/l;  $[Fe^{+3}] = 0,6 \%$ ; pH= 1; temperature= 20° C.

## 5. Conclusions

There were carried out initial studies of gold leaching PCB's from mobile phones discarded as raw material and non-toxic reagents. The employing of these reagents could be the starting point in the development of alternative and ecological processes, in comparison with cyanide processing. Also, it is important to develop processes capable for the treatment of every gold source, either ore or other material with gold contents.

In the ammonium thiosulfate leaching of the PCB's it was determined that gold dissolution is highly dependent of ammonium thiosulfate concentration. With a concentration of 0,13 M it was achieved a dissolution performance around the 70 % in a short leaching time of two hours.

Gold dissolution using acid thiourea solutions reported a performance of 40% with a thiourea concentration of 24 g/l.

According with the conditions studied in the leaching tests, ammonium thiosulfate was more efficient than thiourea in gold dissolution. However, both reagents have a performance lower than expected.

In order to increase gold dissolution rates to values over the 90 %, it is necessary to study the effect of varying other parameters such as cupric ion concentration, pH, temperature, leaching time and solid-liquid ratio in ammonium thiosulfate leaching, and in the other case, ferric ion concentration, pH and temperature.

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Department of Metallurgical Engineering, National Technological University, Regional Faculty of Córdoba. Department of Materials and Technology, Faculty of Exact, Physicals and Natural Sciences, National University of Córdoba . Mining Lab, Ministry of Mining, Industry and Commerce of Córdoba.

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