

Gold Separation of Handphone Circuit Board Scraps (PCBs) by Leaching Process

Yusnimar Sahan,^{a,*} and Drastinawati,^a

^{a)} *Chemical Engineering, Universitas Riau, Indonesia*

* Corresponding author: yusnisahan@lecturer.unri.ac.id

Paper History

Received: 27-June-2016

Received in revised form: 10-July-2016

Accepted: 30-July-2016

H_2SO_4 Sulphouric acid
 HNO_3 Nitric Acid
 $NaOH$ Sodium Hydroxide

ABSTRACT

This study aims to separate the gold of handphone circuit board scraps (PCBs) using $Na_2S_2O_5$ methods that are environmentally friendly. The paper reports a promising method to gold separation of PCBs. The gold separation consisted of three phases; the determination of the optimum conditions using the method of $Na_2S_2O_5$ solution of the gold standard and the application of methods of $Na_2S_2O_5$ in used electronic scraps. Characterization of gold was analyzed by XRF and AAS. Hydrothermal process was applied, aqua regia and HNO_3 solutions were used in the leaching processes, then $Na_2S_2O_5$ as a precipitant in the separation process. Silver, Copper, ferrous, aluminium, lead were solved in HNO_3 and, but gold still as a solid residue. Gold separation of solid residue by leaching with aqua regia. Gold formed as a brown solid with the amount precipitated 0.046g for 5g PCBs samples treated, also it found high purity 99.3%.

KEY WORDS: *Electronic Scraps, Gold, Separation.*

NOMENCLATURE

Au Aurum
 Ag Silvert
 AAS Atomic Absorption Spectrophotometer
 HCl Hydrochloric acid

1.0 INTRODUCTION

Gold, a precious metal with two important characteristics, it has zero resistance to corrosion and for many years, it has been used in hundreds of industrial applications. In the electronic and telecommunication industries, gold has been applied for signal strength than copper [1]. Gold content recovered from computer circuit main board, printed circuit board, handphone printed circuit board (PCBs) [2, 3, 4 & 5].

Sodium bisulphite used as primary and secondary for replacement is important as gold extractor. Sodium bisulphite can separate the gold, forming a complex compound of ionic. The reaction is carried out repeated addition of bisulphite, and the results that can be obtained is 99% of the gold can be extracted [6]. The equation as follows:



Usage limits the amount of reagent on the relative reaction, it is stable in acidic conditions (pH 1 - 2) and neutral pH, and it quickly decomposes in an alkaline solution or alkaline. Sample was leaching easily by using reagents at pH 1 - 2, optimization can be done under away of adjusting the pH, potential's reduction-oxidation, the concentration of sodium bisulphite and leaching time. Pyper and Hendrix (1981) suggested that leaching velocity is depends on the interaction between sodium bisulphite and oxidant, and the use of ferric ions in the sulfuric acid is the most effective way, also ferric ion bound together by a bisulphite, forming complex compounds iron- bisulphite, Separation of gold is very dependent on pH condition. Sodium bisulphite has several advantages than cyanide such as low

sensitivity to metals (Pb, Cu, Zn), and the residual sulfur in the calcine; high gold content in the pyrite and chalcopyrite, and some carbonaceous [7, 8].

In addition, the toxic level of Sodium bisulphite is lower than cyanide [9, 10, 11, 12], and it better able to separate gold from the cyanide. If using cyanide solution (acidic) to be neutralized in advance by Cyanidation, while sodium bisulphite may be used directly [13, 14]. In this research, gold separation of phone circuit boards (PCBs) has been done by leaching and precipitation methods which modified from methods already existence [3, 6 & 7]. Recovery gold of electronic scraps has been done by applied pyrometallurgy and hydrometallurgy methods [3, 5, 6, 7, 8, 9, 10, 11 & 12].

2.0 METHODS

2.1 Materials

Materials are used HNO₃, H₂SO₄, HCl, NaOH, Na₂S₂O₅, Aqua demineralisation (aqua DM) and diethyl malonate. Some of the materials are commercial grade and other analytical grade. These materials were purchased from a local chemical supplier Brataco-Pekanbaru.

2.2 Gold Separation Process

This research has been done using several steps namely sample preparation, leaching processes, gold precipitation and gold purified. Gold content of sample is determined by Atomic Absorption Spectrophotometer (AAS) Shimadzu AA 7000 with method SMHW for standard solution preparation (Figure 1)

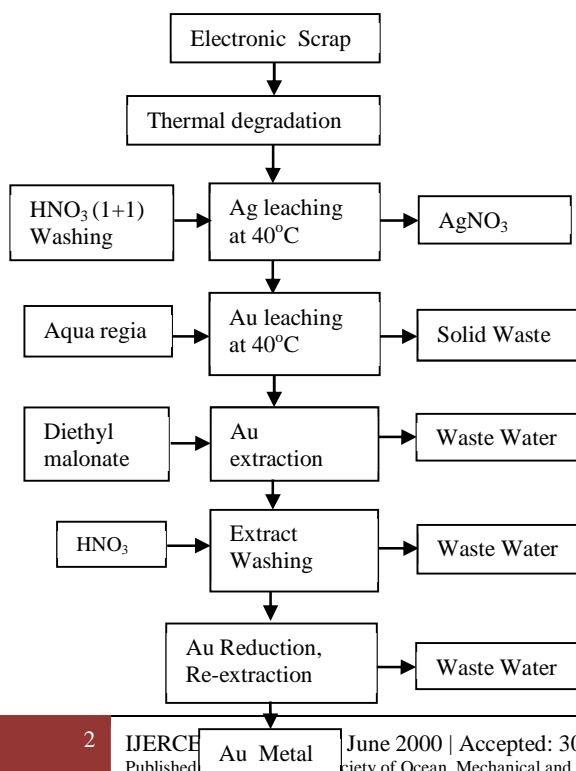
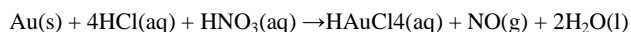


Figure 1: Gold separation process by using leaching process

Handphone Circuit boards (PCBs) is screening into small particle sizes. This particle was analyzed by XRF (Philips Magix) and AAS to determine the metals contents of sample. Sample 5g was leaching with HNO₃ while it heated at 40°C, then the solid residue is separated from filtrate. The solid residue is leaching with aqua regia (30 ml HCl : 3 ml HNO₃) under condition ratio solid to leachant (1:3) and it is heated at 40°C. Afterward, in this mixture Au is extracted by using with diethyl malonate as a solvent. The extractant is reacted with H₂SO₄ + H₂O₂ + (COOH)₂ until brown solid occurring. This solid is separated from mixture, in order to get Au metals then it is purified by burning it with oxy-butane gases.

Reaction identification for Cu(II), Ni(II), Pb(II), Al (III), Fe(III), and Zn (II) ionics has been done in the filtrate. Then, filtrate is added with 7 ml H₂SO₄, and it heated at 145 °C until it is volume remaining 5 ml. Afterward, it is added with 3 ml HCl and heated until it is volume remaining 4 ml, then it is diluted with aqua DM until it's volume 50 ml. In this solution gold is formed as an ionic complex [AuCl₄], then it is reacted with sodium metabisulphite (Na₂S₂O₅) to get Au solid in a mixture solution.

For preparation a [HAuCl₄] 1000 ppm solutions has done by destruction of gold metals with aqua regia then diluted it with aqua DM. The equation reaction:



A series [HAuCl₄] solutions namely 100, 200, 300, 400, dan 500 ppm were prepared, then the absorbance was measured respectively on AAS at 242.8 nm wavelength. The curve calibration standard was done based on these data in order to determine the Au concentration in PCBs

3.0 RESULTS AND DISCUSSION

Handpone circuit boards scraps (PCBs) contains many precious metals such as gold (Au), silver (Ag), copper (Cu), nickel (Ni), lead (Pb), Aluminium (Al), ferrous (Fe), zinc (Zn). Result, based on AAS and XRF analysis of several metals in the PCBs as shown in Table 1.

Table 1: Results Analysis of the PCBs

Component	Amount
Au	85 ± 0.5 ppm*
Ag	0.08%
CuO	2.46%
NiO	0.22%

PbO	0.53%
Al ₂ O ₃	1.74%
Fe ₂ O ₃	0.43%
ZnO	0.15%
Others	-

*AAS Analysis result

Based on AAS analyzed results, the precious metal of Au found in PCBs approximately 85 ± 0.5 ppm. Saadatjoo N., et. al. (2013 also found Au content of the PCBs was around 90 ppm maximally before Au separated from handphone circuit board scraps (PCBs). Furthermore, based on XRF analyzed result the other metal such as copper as CuO was found as the highest amount of metals than among metals in PCBs, following by aluminium, lead, ferrous, nickel, zinc, and silver (Figure 2).

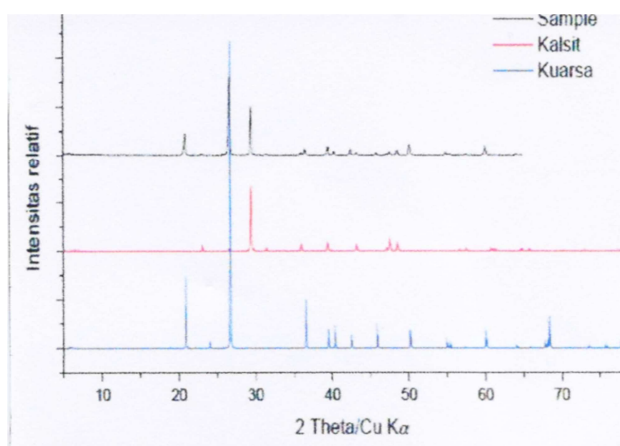
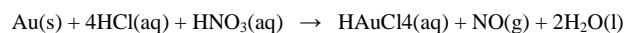
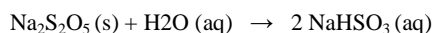


Figure 2: Diffractogram of PCBs sample

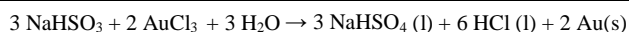
Gold separation from other metals has been done using HNO₃ as a solvent in leaching process, gold in the solid residues and others metals in the filtrate. Then Solid residue is separated from filtrate. Gold of solid residue is leaching with aqua regia and it turn into as an ionic complex [HAuCl₄].



Based on molecules orbital theory, [AuCl₄] complex is one of point group *D_{4h}* compound with planar square form [1]. The fourth ligands Cl⁻ have equal position of centre atomic [13] Compound [HAuCl₄] complexes is reacted with Na₂S₂O₅. Firstly, Na₂S₂O₅ is reacted with H₂O to produce sodium bisulphite :



Then, sodium bisulphite is reacted with ionic complex [AuCl₄] as a results Au is produced as a solid with reaction equation :



In order to get gold metal of samples, Au ions in mixture is treatment by several methods such as precipitation, solvent extraction, adsorption, and ion-exchange [9&10]. In this research, in order to get a gold metal of sample which treated, Au solid is heated at 1064°C (1947.52°F) by using oxy-butane gas until gold metal is obtained.

Table 2: Comparison Precipitation of gold in different Particle size of samples

Particle size of sample (µm)	Reductant	Temperature & Time Reduction	Au content	Purity (%)
50	Na ₂ S ₂ O ₅	40 °C, 30 min	0.009 g	97.9
100	Na ₂ S ₂ O ₅	40 °C, 25 min	0.011g	97.9
150	Na ₂ S ₂ O ₅	40 °C, 20 min	0.025g	98..5
200	Na ₂ S ₂ O ₅	40 °C, 20 min	0.046g	99.3

As can be seen in Table 2, gold was successfully separated by using precipitation process with Na₂S₂O₅. Amount of Au content after reduction is high and can be obtained in short time under the same temperature Reductant (°C). Young et al. (2009) developed a process for separating gold from used goods, by dissolving 70g sample in aqua regia n 1L (pd temperature 90-1000°C) using titanium reactor. After the sample soluble (liquor), Au separated by extraction using dybutyl karbitor (DBC) as extractant. Optimization of extraction, Au move / in the DBC 190g / L (DNG total efficiency of 99%, the ratio liquor / DBC 10: 1), whereas in the rest of Dlm kada Au liquor 30mg / L. The amount of ions Pt, Pd, Ag, Cu and Zn were joined extracted into the phase of DBC is only small (minor). While the allowance Fe pd liquor done by adjusting the pH liquor be pH 3, prior to extraction. Au separation of DBC done with 25% ammonia, precipitation of compounds Au₂O₃·3NH₃, then Au separated from DBC DNG way of precipitation. Then the precipitate is reacted with hydrazine in order to obtain the gold powder with a purity level of 99.9998% (> 5N).

Young and Derek (2009) to separate the gold from printed circuit boards (WPCB), using aqua regia as a solvent (leachant), the ratio between the sample and aqua regia is 1/20 (g/ml), the obtained mixture. Solids separated from the liquid phase of the mixture pd. The precipitate was washed 2.5 times with demineralized water, then Au separated toluene extraction DNG as ekstraktant and tetraoctylammonium bromide as a phase-transfer reagent (10 mM in toluene). The ratio between extractant organic (OE) and aqueous solution (AS) affects the selectivity of gold extraction in the mixture, then the solution (100 ml leachant) mixed with ekstraktant (50), 100 and 200 ml, and the two-phase mixing stirring speed of 400 rpm in 10 minutes and added with 50 mg Dodecanethiol to the organic phase. Sodium borohydride is also added slowly with the intention of preventing the reduction of metal ions in the aqueous solution. After stirring for 2 hours, the organic extractant, toluene, separated, evaporated

to 10 ml in an evaporator and mixed with 250 ml of ethanol for the thiol aside. The mixture is cooled for 5 hours in a temperature of 20°C and a brown precipitate is filtered and washed again with ethanol 97 wt% gold had been separated in the form of nanoparticles.

Zhou et al. (2005) Patented gold separation process on the electronic waste (a mixture of metal and plastic). Garbage is heated in a temperature of 400-500°C for 8-12 hours to melt / burn the plastic material, and the residue is a crude metal. Crude metal / residue reacted with HCl or H₂SO₄ at 90°C to separate the pure metal material, then separated between sediment / solids with liquids. Thereafter, the precipitate was dissolved by HNO₃ (with ratio 1: 2) at a temperature of 60°C to separate silver from other metals. The last process, HCl and NaClO₃ used for leaching gold, gold recovery results are to be obtained in this way achieve > 92%.

Kinoshita et al. (2003) examined the gold separation process in a non-mounted cable circuit (PWBs). At first PWBs is cut into small pieces (± 22 mm), and HNO₃ solvent used in the separation process. Elements of base metals (Cu and Ni) were dissolved with HNO₃ and gold obtained from the separation process in the form of pure pieces. Copper and nickel separation process is done through two stages, leaching and solvent extraction. In the previous process, after Ni separately using HNO₃ solvent 0.1 M, increased levels of 1.0 M HNO₃ acid in order to dissolve the remnants of metal Ni and Cu in the sample. So that the metal gold can be separated perfectly in the form of small fragments. The degree of purity of gold that can be obtained is 99%.

4.0 CONCLUSION

Handphone circuit board scraps (PCBs) contains gold, silver, copper, nickel, lead, ferrous, aluminium and zinc metals. The precious gold metal of PCBs has been separated by a promising method. Aqua regia and HNO₃ solvents were used in the leaching processes, and Na₂S₂O₅ was applied in a precipitation process. For 5g PCBs samples is treated, gold formed a brown solid with the amount precipitated is 0.046 g, also gold found high purity 99.3%.

ACKNOWLEDGEMENTS

This research funded by *Universitas Riau* through Hibah Bersaing DIPA-UNRI in 2015. Thanks for sponsors and all providing support also suggestions for this research.

REFERENCE

1. Lide, D. R., ed. (2005). *CRC Handbook of Chemistry and Physics* (ed. 86th). Boca Raton (FL): CRC Press. ISBN 0-8493-0486-5
2. Widmer, R., Oswald-Krapf, H., Sinha-Khetriwal, D., Schnellman, M., Boni, H., (2005). *Global perspective on E-waste*. Environ, Impact Asses. Vol. 25, pp: 436–458.

3. Oleszek, S., Grabda M., Shibata E. & Nakamura T., (2013). *Distribution of Copper, Silver and Gold during Thermal Treatment with Brominated Flame Retardants.*, Waste management. Vol. 13, pp: 1835-1842, Journal homepage; www.elsevier.com/locate/wasmen
4. Montero, R., Guevara, A., De La Torro E., (2012). *Recovery of gold, silver, copper and Niobium from printed circuit boards using leaching column*. Journal of Earth Science and Engineering. Vol. 2, pp: 590-595.
5. Kinoshita, A., Alkita, S., Khobayashib N., Niib S., Kawazumih F., Takhashi K., (2003). *Metal recovery from non-mounted printed wiring boards via hydrometallurgical Processing*. Hydrometallurgy, 69, pp: 73-79.
6. Syed, S., (2012). *Recovery of Gold from Secondary Sources-A review*, Hydrometallurgy 115-116; pp: 30-51, Journal homepage; www.elsevier.com/locate/Hydromet.
7. Park, Y.J. & Fray, D.J. (2008). *Recovery of High Purity Precious Metals from Printed Circuit Boards*. Journal of Hazardous Materials. Vol. 164, pp: 152-1158.
8. Cui, L., Zhang, L., (2008). *Metallurgical recovery of metals from electronic waste; a review*, J. Hazard Mater. Vol. 158, pp: 228–256.
9. Quinet P., Proost, J., Van A.L., (2005). *Recovery of precious metals from electronic scrap by hydrometallurgical processing routes*. Min, Metals. Process, 22 (1), pp: 17-22.
10. Byoung J.K., Jeon W.A., Seong J.K., Tam T., Myong J.K., (2009). *Processing high purity gold from scraps using diethylene glycol di N-butyl ether (dibutyl carbitol)*. Hydrometallurgy 95, pp: 262-266.
11. Zhang, S., Forssberg, E., (1997). *Electronic scrap characterization for materials recycling*. J Waste Manage. Resource, Recov. 3, pp:157–167.
12. Saadatjoo N., Heydari H., Abdullah A., Behzadi, M. (2013). *Recovery of gold from computer circuit board scraps: the study of the effect of different reductants*. Journal of Applied Chemistry. Vol 8, N0.27, pp: 55–60.
13. Tuncuk, A., Stazi, V., Akcil, A., Yacizi, E.Y., Devenci, H., 2012. *Aqueous metal recovery techniques from e-scrap; hydrometallurgy in recycling*, Minet. Eng. 25, pp: 28–37.