Gold Leaching from Printed Circuit Boards (PCBs) as one of the Urban Mine Resources using Thiosulphate: Optimization using Response Surface Methodology (RSM)

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Abstract. Along with the decrease of metal resources in the natural ores, urban mine comes up as an alternative with two aspects of both resource and environmental protection due to recycle. Broken printed circuit boards (PCBs) as one of the urban mine resources contains valuable metals, such as, Au, Ag, and Ti, with high concentration, however, it has not been seriously managed in some developing countries, such as Indonesia. Focusing on gold extraction, safer leaching agent using thiosulphate has been applied in this study and scrutinized the optimum leaching conditions with respond surface methodology. Temperature and concentration of thiosulphate were varied and designed in accordance with three factorial. Leaching time was set for two hours and the shaking speed of waterbath was set at fixed value of 200 rpm. In order to quantify the final concentration of gold in the solution, Atomic Absorption Spectroscopy (AAS) was applied for all samples. Response Surface Method (RSM) is utilized to know the optimum result of Au extracted. The result shows that the optimum result of Au extracted by using ammonium thiosulphate is 0.5114 mg/L respectively with correlative equation as follow;

Gold Concentration = 0,127+0,00268*T+0,66*C-0,000021*T^2+1,74*C^2-0,0038*T*C.

With T = temperature (°C) and C = thiosulphate concentration (M)

The obtained R^2 is 0.94 and the p-value is 0.037 showing the significance of the variable correlation in the equation.

Keywords: PCB, Gold extraction, thiosulphate, optimization, RSM.

1. Introduction

Nowadays, the development of technology is running very fast which causes the latest devices to quickly become obsolete. As for impact for this situation, there are 50 million tons of electronic scrap resulted wordwidely every year. Unfortunately, some developing countries do not have appropriate regulations regarding on how to manage electronic scrap which are classified as hazardous waste [1,2].

Printed circuit boards (PCBs) are the main integrated components of electronic devices. As it is shown in literature data, about 70% of the PCBs composition consists of ceramic and plastic material, 16-20% of copper, 4-5% of solder, 1-3% of zinc, 2-3% of iron, 1-2% of nickel, 600-1000 ppm of Ag, 150-300 ppm of Au and 80-150 ppm of Pd [3,4]. PCBs contain heavy metals that are associated with health and environmental problems. 3% of the electronic scrap produced annually in various parts of the world is PCBs [5].
One source of PCBs is mobile phones. In 2017, mobile phones use in Indonesia is amounted to 25% of the total population of Indonesia which has reached 262 million inhabitants. The use of mobile phones in Indonesia is a large potential for PCBs waste in the future, but it is also beneficial because it contains gold which is precious metal [3]. A solution that would overcome the existence and abundance of hazardous waste and obtain the metals without mining the natural ore. This kind of resource is classified as urban mining.

In general, gold recovery process can be carried out using pyrometallurgical or hydrometallurgical methods. Utilization of high temperature of pyrometallurgical process can easily cause serious environmental problems, high energy and cost. Hydrometallurgy can offer an effective solution due to its flexibility, environmental friendly and energy saving features.

That is why in this study, PCBs waste is leached using reagent which is less harmful to the environment.

Aqua regia as well as sodium cyanide are the solution which is usually applied for gold extraction with high gold dissolution rate, however, there is potential risk for environmental damage. This is due to the risk presented by the solution for the health and safety risks of the workers, not to mention the serious threat for the environment [6]. An alternative approach is to use thiosulfate to leach gold from the PCBs waste. Thiosulfate is considered as nontoxic reagent in gold leaching, an alternative to cyanide, that has gold leaching capability faster than cyanide [7].

Leaching of gold from PCBs using ammonium thiosulfate has been carried out by several researchers [8,9]. However, until now, there has no information reported on the optimization of the leaching process of gold from PCBs using ammonium thiosulfate. In this research, leaching optimization was conducted using response surface methods (RSM) with temperature and concentration of ammonium thiosulfate as independent variables.

2. Research Methodology

2.1 Material

The initial step that is conducted was to separate gold containing part from the PCBs waste. After that, the gold containing parts were crushed and sifted to pass the 25 mesh sieving. The chemical analysis of sample was done using EDX as shown in Table 1.

| Elements | Weight (w/w, %) | Elements | Weight (w/w, %) |
|----------|----------------|----------|----------------|
| Cu       | 34.812         | Ti       | 0.927          |
| Si       | 21.169         | Pb       | 0.529          |
| Ca       | 10.713         | Cr       | 0.328          |
| Br       | 9.529          | Sb       | 0.308          |
| Ni       | 6.590          | Ag       | 0.275          |
| Sn       | 3.919          | Sr       | 0.190          |
| Zn       | 3.333          | K        | 0.176          |
| Ba       | 2.472          | Mn       | 0.055          |
| Au       | 2.105          | Zr       | 0.043          |
| Fe       | 1.265          | Nb       | 0.017          |
| S        | 1.247          |          |                |

Based on the information of chemical composition of raw material as shown in Table 1, the gold composition in the sample is 2.105% wt. This amount is quite high comparing to the gold natural ore which is usually in the concentration of 5-10 ppm.

2.2 Methodology
Six gram of sample was leached in ammonium thiosulfate at various concentrations (0.1 M; 0.2 M and 0.3 M) using silicon covered erlenmeyer. Leaching experiment was carried out in a water-bath shaker with agitation speed of 200 rpm and various temperatures (30 ˚C, 60 ˚C and 90 ˚C) for 2 hours. Sampling was conducted from the suspended solution by separating the solid phase from the aqueous using centrifuge. In order to determine the gold concentration in the filtrate or aqueous phase, atomic absorption spectrophotometry (AAS) was utilized.

Further optimization study was designed using response surface methods (RSM), in order to justify the operating conditions to leach Au from PCBs waste. The design response was carried out using minitab 18 with concentration of Au (Y) as the dependant variable, while the independent variables consist of ammonium thiosulfate concentration (X1) and the temperature (X2).

3. Results and Discussion
3.1 Effect of ammonium thiosulfate concentration and temperature on the leaching behaviour of Au from PCBs waste
The effect of two independent variables used in this study are shown in Figure 1. From figure 1, it can be seen that the concentration of Au is getting higher when high ammonium thiosulfate concentration was applied. While, as for temperature, the concentration of Au does not change significantly. There is tendency of lower Au concentration extracted when high temperature was applied. It shows that ammonium thiosulfate concentration has much better significance than temperature as operating condition. Steep curve at the ammonium thiosulfate concentration changes, as shown in Figure 1 confirmed this phenomenon and in accordance with the kinetics. To determine effect of two independent variables on the dependent variable, the data has been analysed using response contour plot as shown in Figure 2. Based on Figure 2, it can be seen that the maximum response value is > 0.5 mg/L of Au concentration in the thiosulfate solution. This condition was obtained with working temperature in between 30 - 60˚C and ammonium thiosulfate concentration of > 0.29 M. From the RSM, equation 1 can be generated to show the relationship of variables as follows :

$$Y = 0.127 + 0.66 X_1 + 0.00268 X_2 + 1.74 X_1^2 - 0.000021 X_2^2 - 0.00380 X_1 X_2 \ldots \ldots \ldots (1)$$

Using statistic analysis, it can be justified that equation 1 is in a good agreement with the experimental data with the R² value of 0.95.
Figure 1. Main effects plot for Au Concentration

Figure 2. Contour plot the difference effect of ammonium thiosulfate and temperature condition on Au concentration

3.2 Optimization of the Au concentration
The purpose of this optimization procedure is to minimize the required effort or operational cost and maximize the desired product which is Au extracted from PCBs waste. Figure 3 shows
the optimum point in the leaching of Au from PCBs waste [10]. The optimum point occurs at 37.27 °C and the ammonium thiosulfate concentration of 0.30 M. Based on Figure 1 and 3, it can be seen that the higher of ammonium thiosulfate concentration, the higher concentration of Au produced. This is consistent with the theory that the higher concentration of acid makes the leaching rate increase. While the higher temperature, the lower of Au concentration obtained. Based on these data, it is necessary to study the thermodynamic relationship of this reaction whether the reaction runs in an endothermic or exothermic reaction. Several studies have been carried out the gold leaching at ambient temperature [3,9,11,12].

Figure 3. Optimal design of leaching Au from PCB’s waste

4. Conclusion
The variable that significantly influences to the leaching of Au from PCBs waste is the ammonium thiosulfate concentration. The optimal conditions in the leaching of Au from PCBs waste: ratio S/L of 6/100, stirring speed of 200 rpm, temperature at 37.27 °C, ammonium thiosulfate concentration of 0.3 M and the period of leaching for 2 hours.

References
[1] Petter, P.M.H., Veit, H.M., and Bernardes, A.M. 2014. Evaluation of gold and silver leaching from printed circuit board of cellphones. Waste Management, 34(2), pp. 475-482.
[2] Chaurasia, A., Singh, K. K., and Mankhand, T. R. 2013. Extraction of Tin and Copper by Acid Leaching of PCBs. International Journal of Metallurgical Engineering, 2(2), pp. 243–248.
[3] Birloaga, I. and Vegliò, F. 2016. Journal of Environmental Chemical Engineering Study of multi-step hydrometallurgical methods to extract the valuable content of gold, silver and copper from waste printed circuit boards. Biochemical Pharmacology. Elsevier B.V., 4(1), pp. 20–29.
[4] Kan, Y., Yue, Q., Gao, B., Li, Q., 2015. Preparation of epoxy resin-based activated carbons from waste printed circuit boards by steam activation. Mater Lett. 159, 443-446.
[5] Ghosh, B. et al. 2015. Waste Printed Circuit Boards recycling: An extensive assessment of current status. Journal of Cleaner Production. Elsevier Ltd, 94, pp. 5–19.
[6] Geoffroy, N. and Cardarelli, F. 2005. A method for leaching or dissolving gold from ores or precious metal scrap. The Journal of the Minerals, Metals and Materials Society. 57 (8), pp. 47-50.
[7] Aylmore, M., and Muir, D. 2001. Thiosulfate leaching of gold – a review. Minerals Engineering. 14 (2), pp. 135-174.
[8] Camelino, S., Rao, J., Padilla, R.L., and Lucci, R. 2015. Initial studies about gold leaching from printed circuit boards (PCB’s) of waste cell phones. Procedia Materials Science 9, pp. 105-112.
[9] Tripalhi, A., Kumar, M., Sau, D.C., Agrawal, A., Chakravarty, S., and Mankhand, T.R. 2012. Leaching of gold from the waste mobile phone printed circuit boards (PCBs) with ammonium thiosulphate. International Journal of Metallurgical Engineering, 1(2), pp. 17-21.
[10] Ferdana, A.D., Petrus, H.T.B.M., Bendiyasa, I.M., Prijambada, I.D., Hamada, F., and Sachiko, T. 2018. Study on Sumbawa gold recovery using centrifuge. IOP Conference Series: Materials Science and Engineering. Volume 285, Issue 1, Pages 012027.
[11] Baghalha, M. 2007. Leaching of an oxide gold ore with chloride/hypochlorite solutions. International Journal of Mineral Processing, Volume 82, pp. 178-186.
[12] Bisceglie, F., Civati, D., Bonati, B., Faraci, F.D. 2017. Reduction of potassium cyanide usage in a consolidated industrial process for gold recovery from wastes and scraps. Journal of Cleaner Production. Volume 142, pp. 1810-1818.

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