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Optimization of the leaching process of the printed circuit boards production's sludge for copper recovery via electrolysis

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Abstract

Copper is the main component of printed circuit boards (PCBs) which is the main part of an electronic device. The manufacture of PCBs often releases huge amount of sludge which contained copper. Derived from the demand of the industrial park about the wasted sludge treatment and the reducing of the plantation of natural mineral resources, the copper recovery process from PCBs manufacture's wasted sludge was studied. The process of copper leaching by sulfuric acid solution was studied using experimental planning methods using the simulation software Modde 5.0. The parameters were calculated and simulated to evaluate the effect of the acid concentration, leaching time, and the ratio between the solid and liquid phases on the leaching efficiency. The obtained results by simulation were compared with real experimental results. The obtained results from this study are planning in the actual deployment of the Hanoi Urban Environment One Member Limited Company in Vietnam.

Keywords. Printed circuit board, copper sludge, leaching.

1. INTRODUCTION

The production of electronic equipment is rapidly increasing in the world as a result of the revolution of information technology. Consequently, waste electronic equipment is a growing concern globally. Printed circuit boards (PCBs) are used in almost all electronics today and fabricated in large volumes across the world. The increasing volume of waste discharges from PCBs manufacture poses an environmental threat, as this sludge can contain substances that are harmful, such as heavy metals, nitrates and phosphates. Copper is one of the most used materials in the production of PCBs [1]. The copper recovery from sludge of the PCBs production became more important lately because of the continuous increase of copper demand, as well as the urgent in solving the environmental issues.

One of the big challenges for copper recovery via hydrometallurgy from copper sludge is how to form a concentrated copper ion solution and leaching is a popular method to solve this problem. There are some different leaching techniques such as bio-leaching, heap leaching, oxidized leaching, pressure leaching, and also, leaching process can be done using acid or base solutions. Although there are some disadvantages, especially the separation of copper from the other metal contaminants but acid leaching process is still preferred for low-grade ores and solid waste treatment because of the high leaching efficiency. Nitric acid and hydrochloric acid can dissolve low-grade ore or e-waste easily but because of the bad effect of Cl^-, and NO_3^- anions to the next step - electrodeposition, these acids are not normally used in the real industrial techniques [2,3]. Therefore, cause of the conformity of the copper sulphate solution for electrowinning, the leaching process using sulphuric acid has widely been used for the copper recovery from low-grade complex ores [4, 5] or industrial wastes [6].

In this paper, the leaching process of the copper sludge from PCBs production has been carried out using statistical design of experiments for different process parameters. The real experiment's and simulation' results were compared. The response surface modeling has been performed to optimize the best conditions for leaching of copper from the sludge by using Modde 5.0 software on a personal computer.

2. EXPERIMENTAL

2.1. Materials and reagents

PCBs wastes used in this study were sourced from Hanoi Urban Environment One Member

Limited Company (Urenco). The desiccate PCBs sludge was grinded and sieved to achieve the size smaller than 0.1mm. Analytical grade sulfuric acid was used in the experiments. The leach solutions were prepared by dissolving the desired volume of sulfuric acid in deionized water to the required concentration.

2.2. Leaching process

The experiments were conducted at room temperature in a 0.5 L glass beaker, using a magnetic stirrer rotating at a speed of 600 rpm. In each experiment, a quantity of 100 mL sulfuric acid was introduced into the reactor. When the desired stirring speed was reached, the solid sample was added into the reactor. The amount of solid sample was changed to satisfy the ratio of solid/liquid. After specific time intervals, the leaching solutions were filtered. The copper concentration in the filtrate was determined using UV-Vis spectrophotometry at the wavelength of 805 nm. The leaching efficiency was calculated based on the copper concentration in the sample before and after leaching.

2.3. Modeling and Simulation

Method of modeling and statistical design of experiments on quadratic central composite design (CCD) improved by Box and Hunter [7] was used to model the surface response in this study. The leaching efficiency was taken as the response variable, where the concentration of sulfuric acid (X_1, M), time of leaching (X_2, min) and the ratio of solid/liquid (X_3, %) were considered as the independent variables (factors). The relationship between the coded values and the experimental values was described by the following Eq. (1).

$$x_j = \frac{Z_j - Z_j^0}{\Delta Z_j} \qquad (1)$$

where x_j, Z_j are the coded and the experimental value of the independent variable respectively, Z_j^0 is the experimental value of the independent variable at the center point level, and ΔZ_j is the step change of variable.

Table 1: Central composite rotatable design arrangement and results

Run	Independent variable			Eff.
	$[H_2SO_4]$ (M)	Leaching time (min)	The ratio of S/L (%)	
1	0.8	30	10	0.77
2	1.2	30	10	0.91
3	0.8	90	10	0.80
4	1.2	90	10	0.95
5	0.8	30	14	0.75
6	1.2	30	14	0.81
7	0.8	90	14	0.78
8	1.2	90	14	0.88
9	0.66	60	12	0.66
10	1.34	60	12	0.85
11	1.0	9.54	12	0.89
12	1.0	110.46	12	0.96
13	1.0	60	8.36	0.88
14	1.0	60	15.36	0.79
15	1.0	60	12	0.96
16	1.0	60	12	0.95
17	1.0	60	12	0.93
18	1.0	60	12	0.94
19	1.0	60	12	0.93
20	1.0	60	12	0.95

Table 1 shows the total number of required experimental runs and the levels in coded and actual value of the acid concentration. The leaching time and solid/liquid ration were shown in table 2.

Table 2: Independent variables and their corresponding levels

Independent variables	Symbol code	Code variable levels				
		-1.682	-1	0	1	1.682
Sulfuric acid concentration (M)	Z_1	0.6636	0.8	1.0	1.2	1.3364
Time of leaching (min)	Z_2	9.54	30	60	90	110.46
The ratio of solid/ liquid (%)	Z_3	8.36	10	12	14	115.36

The results of 20 trials were also entered into the Modde 5.0 software in order to fit the model by multiple linear regressions. The previous investigation indicates that the best-fit model of response Y is quadratic model. This model is given by the general expression:

$$Y = b_0 + \sum_{i=1}^{k} b_i X_i + \sum_{i=1}^{k} b_{ii} X_i^2 + \sum_{i,j=1(i\neq1)}^{k} b_{ij} X_i X_j$$

where Y is copper leaching efficiency, X_i, X_j, are the coded values of independent variables, the terms X_iX_j and X_i^2 represent the interaction and quadratic terms, respectively, b_0 is the constant coefficient, b_i, b_{ii}, and b_{ij} namely the linear, quadratic and interaction coefficient.

3. RESULTS AND DISCUSSION

3.1. Modeling of copper leaching efficiency

Table 3 shows the estimated regression coefficients for the leaching efficiency model. The confidence level of model was 95 %. The model was then verified by checking the Student's t-distribution. At the confidence level P = 0.05, the coefficients b_{22}, b_{21}, b_{23} did not satisfy and hence they were rejected. The accuracy and variability of the above model could also be evaluated by the coefficient of determination (R^2). The R^2 for the copper leaching efficiency was calculated to be 0.971, explaining that the variability of response is at 97.1 % confidence level, and only 2.9 % of the total variations cannot be explained by the model. Moreover, the value of adjusted determination coefficient (adj. R^2) of 0.945 was also close to 1. Therefore, the predicted model for copper leaching efficiency had a good agreement with the experimental data.

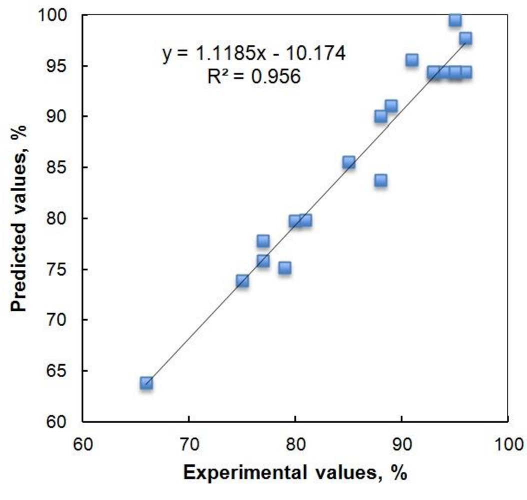

Figure 1: Linear correlation between observed and predicted values for copper leaching

Final regression equation for the copper leaching efficiency, which incorporates the types of coded coefficients, is shown in Eq. (2):

$$Y = 0.943 + 0.064X_1 + 0.020X_2 - 0.044X_3 -0.070X_1^2 - 0.042X_3^2 - 0.034X_1.X_3 \quad (2)$$

The model equation indicates that the copper leaching efficiency involves the second and no higher order of sulfuric acid concentration and ratio of solid/liquid. Figure 1 shows plot of the predicted vs. experimental value for copper leaching efficiency. It could be seen that the experimental results were distributed relatively near to a straight line, which confirms the good agreement between the predicted and actual results. This illustrates that the fitted regression coefficient to the equation and the CCD model with an experimental design can be effectively applied for optimization of copper leaching efficiency.

Table 3: Estimated regression coefficients for quadratic model

Source	Coefficient	Standard error	P
Model	0.943	0.001	<0.001
X_1	0.064	0.006	<0.001
X_2	0.020	0.006	0.012
X_3	-0.004	0.006	<0.001
X_1^2	-0.070	0.006	<0.001
X_2^2	-0.014	0.006	0.054
X_3^2	-0.042	0.006	<0.001
X_1X_2	-0.006	0.008	0.454
X_1X_3	0.034	0.008	0.002
X_2X_3	0.014	0.008	0.132

3.2. The interaction among factors and its effect on copper leaching efficiency

Figures 2, 3 and 4 illustrate the interaction among process factors and their effect on copper leaching efficiency.

It can be seen from figure 2 that the shape of the plot was almost uniform when the ratio of solid/liquid and the time of leaching were changed simultaneously. A similar trend was also recorded in figures 3 and 4. This seems to be confirmed that the interaction between each pair of factor is completely independent.

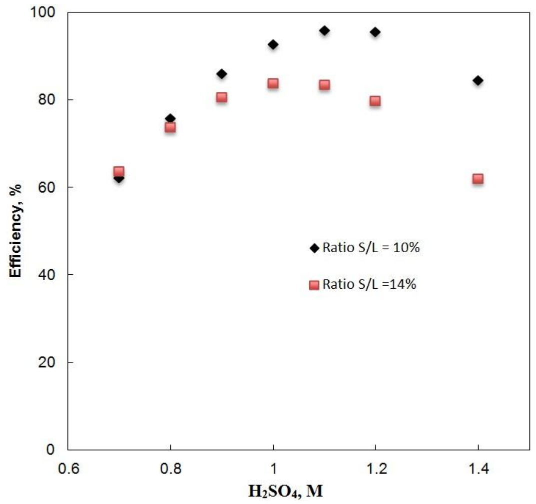

Figure 2: Effect of sulfuric acid concentration and the ratio of solid/liquid on leaching efficiency

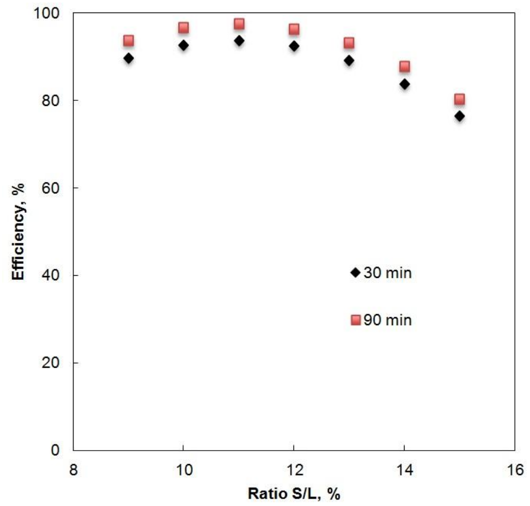

Figure 3: Effect of the leaching time and the ratio of solid/liquid on leaching efficiency

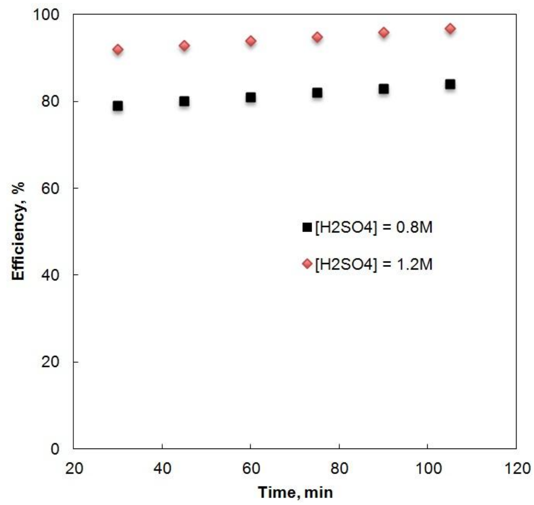

Figure 4: Effect of sulfuric acid concentration and the leaching time on leaching efficiency

3.3. Optimization of copper leaching conditions

In order to find the optimum conditions of copper leaching with the highest efficiency, the lowest consumption of materials and time is stressed upon this investigation. The response surface methodology (RSM) can be used to find desirable location in the design space. While using the RSM with the help of Modde 5.0 software the best conditions of copper leaching process were determined. Table 4 demonstrated the results of the process optimization and optimum levels of variables.

Table 4: Results of process optimization and optimum levels of variables

Proposed range	$[H_2SO_4]$ (M)	0.8-1.2
	Time (min)	30-90
	Ratio of S/L (%)	10-14
Target		Maximum
Optimum values of variables	$[H_2SO_4]$ (M)	1.08
	Time (min)	75.15
	Ratio of S/L (%)	12.59
Leaching efficiency (%)		96.67

The optimum conditions have been found as: sulfuric acid concentration of 1.08 M, leaching time of 75.15 minute, the ratio of solid/liquid of 12.59 %. Under these conditions, the leaching efficiency reached 96.67 %.

4. CONCLUSION

A quadratic model of copper leaching efficiency has been obtained by using RSM based on CCD with the help of Modde 5.0 software. A good agreement between model and experiment is confirmed. The final quadratic model was used to find the optimum conditions for copper leaching, which turn out as sulfuric acid concentration of 1.08 M, leaching time of 75.15 minute, the ratio of solid/liquid of 12.59 %. Maximum value of copper leaching efficiency under optimum conditions was calculated to be 96.67 %.

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