Vietnam Journal of Science and Technology ***56*** *(2C) (2018) 16-22*

THE STUDY ON OPTIMIZATION OF COPPER LEACHING FROM WASTE PCBs BY USING RESPONSE SURFACE METHODOLOGY

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Received: 10 May 2018, Accepted for publication: 20 August 2018

ABSTRACT

Recycling printed circuit boards (PCBs) is an important solution not only to treat hazardous waste but also to recover valuable materials. This research focus on optimizing the leaching copper process from waste PCBs to recycle. This process was performed in $Fe_2(SO_4)_3$ solution at room temperature and using H_2O_2 as an oxidant. For optimization, response surface methodology was used to investigate the different parameters, including $Fe_2(SO_4)_3$ concentration, volume of H_2O_2 addition and leaching time. Design Expert 10.0 software was applied to design the experiments and calculate the regression equation of the response function. As a result, a model was established which is compatible with the experimental data at the correlation coefficient (R^2) of 0.99. The optimal conditions were identified as $Fe_2(SO_4)_3$ concentration of 0.35 M, volume of H_2O_2 addition at 10 mL and leaching time of 10 h. Under this condition, leaching efficiency of 90.5 percent was achieved. After leaching, copper was recovered from extract solution by electrochemical technology with the efficiency of 85 percent. The results from this study hence proposed a very promising method for recycling copper in PCBs.

Keywords: circuit boards (PCBs), leaching, recovery, response surface methodology.

1. INTRODUCTION

Increasing demand for electronic equipments in modern life has been creating pollutional issues and generating 20-50 million tons of electronic wastes (or e-wastes) every year [1]. Because of their hazardous material contents, printed circuit boards (PCBs) is potentially harmful to the environment, human health and the economy [2]. Generally, electronic wastes are composed of metal (40 %), plastic (30 %) and refractory oxides (30 %) [3]. The typical metal scrap consists of copper (20 %), iron (8 %), tin (4 %), nickel (2 %), lead (2 %), zinc (1 %), silver (0.02 %), gold (0.1 %) and palladium (0.005 %) [4]. Increased recycling of electronic waste is supposed to limit the total quantity of waste going to final disposal [2].

PCBs are found in virtually all electric and electronic equipment, provide the electrical interconnections between components [5]. PCBs were recycled in three processes which is pretreatment, physical recycling, and chemical recycling [5]. PCBs recycling generally start from the pretreatment stage, which include disassembly of the reusable and toxic parts and then PCBs are treated using physical recycling or chemical recycling process. Physical processing for the separating the metal fraction and non-metal fraction from waste PCBs includes shape separation, magnetic separation, electric conductivity-based separation, density-based separation and corona electrostatic separation [6]. From the review, we found that chemical recycling methods include pyrolysis, gasification and combustion. Metal fraction can be treated by pyrometallurgical, hydrometallurgical or biotechnological process [5].

Response surface methodology is the most widely used method for experimental design; it can be used to optimize the operating parameters at the minimum experimental runs and analyze the interactions between parameters [7, 8].

Copper occur in nature in directly usable metallic form. Most copper is mined or extracted as copper sulfides from large open pit mines in porphyry copper deposits that contain 0.4 to 1.0 % copper. During mining and refining (purification) of copper, dust and waste gases such as SO_2 are produced which may have a harmful effect on the environment. Recovery of copper is worth up to 90 % of the cost of the original copper [4].

So that, in this paper, copper was chosen to be recycled by leaching process from waste PCBs which contain large amount of copper than another metals. This study used $Fe_2(SO_4)_3$ solution at room temperature to extract copper and H_2O_2 as an oxidant. It is a simple, economic and effective method to recycle copper. Response surface methodology was used to optimize the leaching process to reduce the number of experimental runs and its response surface map was used to locate the optimum response variables in this research.

2. MATERIALS AND METHODOLOGY

2.1. Materials

The material used for this research was collected from Vietnam Australia Environment S.J.Co. (VINAUSEN), Le Minh Xuan Industrial zone, HCM City.

PCBs were crushed to collect powder materials with a particle size below 0.075 mm. Copper concentration in raw material was 15.5 ± 0.8 %. $Fe_2(SO_4)_3$ and H_2O_2 were bought from Merck Chemicals Ltd and distilled water was used throughout the leaching process.

2.2. Extraction process and experimental design

Copper leaching process from waste PCBs was conducted by following the procedure given in Figure 1. PCBs waste containing copper was crushed into powder with the size smaller than 0.075 mm. Raw samples were dissolved in $Fe_2(SO_4)_3$ solution which was then added with certain amounts of H_2O_2 (dropped 1 mL/min) to oxidant. A magnetic stirrer was applied during the experiment. After specific time intervals, the leaching solutions were filtered. $Fe_2(SO_4)_3$ concentration, volume of H_2O_2 addition and leaching time were varied in each experiment. The leaching efficiency was calculated based on the copper concentration in the sample before and after leaching.

Design Expert 10.0 software was used to design the experiments and optimize the leaching process. It is a software for design of experiments which provides statistical tools, such as "two-level factorial screening designs". This software can identify the vital factors that affect the process or product so that it can make necessary improvements.

In the experiment, three key parameters were pre-investigated. Specifically, $Fe_2(SO_4)_3$ concentration changed in range of 0.25 - 0.5 M, amount of H_2O_2 added was from 5 to 15 mL and leaching time increased from 6 to 10 hours. Results were showed that the central values (zero level) chosen for the design were: $Fe_2(SO_4)_3$ concentration at 0.3 M, amount of H_2O_2 at 10 mL and leaching time at 8 hours. These three key parameters and their levels settings are given in Table 1.

Based on the ranges of influence parameters in Table 1, we run Design Expert 10.0 software to have experiment conditions, as given in Table 2.

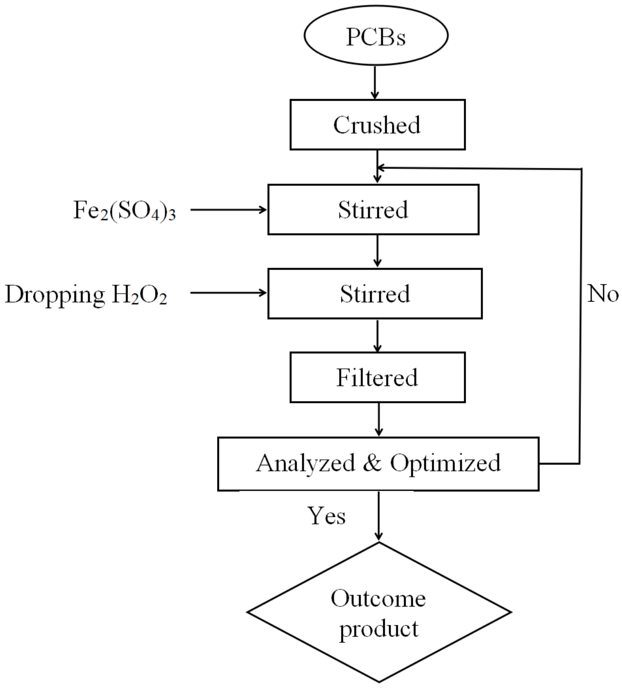

Figure 1. Copper leaching process from PCBs waste.

Table 1. Experimental design.

Independent variables	Symbols	Levels and ranges		
		-1	0	1
Fe^{3+} concentration, M	A	0.25	0.30	0.35
H_2O_2 volume, mL	B	5	10	15
Leaching time, h	C	6	8	10

3. RESULT AND DISCUSSION

3.1. Leaching efficiency

Different parameters, as listed in Table 1, with different matrixes for optimization were input into Design Expert 10.0 software. The software was run and provided different conditions of experimental runs. All experiments were then performed and the leaching efficiency was calculated in each of condition (Table 2). The copper in PCBs was extracted via a redox reaction whereby insoluble copper extracted from waste PCBs was transformed into soluble cupric ion (Cu^{2+}), which in turns was recycled.

Reaction occurs as follows:

$$2Fe^{3+} + Cu \rightarrow Cu^{2+} + 2Fe^{2+}$$

3.2. Response surface analysis

The influence parameters on leaching efficiency was coded: Fe^{3+} concentration (A), H_2O_2 volume (B) and leaching time (C). A typical regression equation from three influence parameters is illustrated as below:

$$Y = b_0 + b_1A + b_2B + b_3C + b_{12}AB + b_{23}BC + b_{13}AC + b_{11}A^2 + b_{22}B^2 + b_{33}C^2$$

The leaching efficiency was calculated in each of experiment and showed in Table 2.

Table 2. Design experiments and results.

Run	**Fe^{3+} concentration, M**	**H_2O_2 volume, mL**	**Leaching time, h**	**Leaching efficiency, %**
1	0.3	5	10	79
2	0.25	15	8	77
3	0.3	15	10	85
4	0.35	10	10	91
5	0.35	5	8	84
6	0.25	5	8	76
7	0.25	10	6	74
8	0.25	10	10	78
9	0.3	15	6	76
10	0.3	10	8	81
11	0.35	15	8	86
12	0.35	10	6	81
13	0.3	10	8	80
14	0.3	10	8	81
15	0.3	5	6	75

To figure out the optimal condition for copper leaching process, data from Table 2 were proceeded to statistically analyze using ANOVA. The regression equation via above coded factors can be used to predict the interactions between influence variables and following responses by given levels of each factor. The ANOVA analysis results are showed in Table 3.

Table 3. Analysis of variance (ANOVA) quadratic model.

Souce	Sum of Squares	Degree of Freedom	Mean Square	F-Value	p-Values Prob>F
Model	303.02	9	33.67	42.98	0.0003
A	171.12	1	171.12	218.46	<0.0001
B	12.50	1	12.50	15.96	0.0104
C	91.13	1	91.13	116.33	0.0001
AB	0.25	1	0.25	0.3191	0.5965
AC	9.00	1	9.00	11.49	0.0195
BC	6.25	1	6.25	7.98	0.0369
A^2	5.03	1	5.03	6.42	0.0523
B^2	4.33	1	4.33	5.53	0.0654
C^2	2.56	1	2.56	3.27	0.1302
Residual	3.92	5	0.78		
Pure Error	0.6667	2	0.33		

Results by Design expert software indicated that the model was statistically significant (P value < 0.001). Factors (A, B, C) and pair of factors (AC, BC) were involved in regression equation (P value < 0.05). All the rest that had p value higher than 0.05, were not considered in the regression equation. The correlation coefficient (R^2) is 0.9872, which is close to 1, indicating that the actual leaching efficiency is consistent with the predicted leaching efficiency. The predicted R^2 of 0.8257 is in reasonable agreement with the adjusted R^2 of 0.9643 (the difference is less than 0.2).

Therefore, the final regression equation can be functionated as below:

$$Y = 80.67 + 4.62A + 1.25B + 3.38C + 1.5AC + 1.25BC + 1.17A^2 - 1.08B^2$$

The equation in terms of coded factors can be used to make predictions about the response for given levels of each factor. This reflected that predicted leaching efficiency of model is linear and similar to the actual leaching (Figure 2). Therefore, predicted leaching efficiency in this model can be reliable to estimate the actual leaching efficiency.

The coefficient estimate represents the expected change in response y per unit change in x when all remaining factors are kept constant. The intercept in an orthogonal design is the overall average response of all the runs. The coefficients are adjustments around that average based on the factor settings. When the factors are orthogonal the variance inflation factors (VIFs) are 1; VIFs greater than 1 indicate multi-colinearity, the higher the VIF the more severe the correlation of factors. As a rough rule, VIFs less than 10 are tolerable.

RSM method showed the interactions between parameters with the minimum experimental runs. From experimental data, values of key parameters were plotted and shown in Figure 3 and Figure 4. The response surface maps indicated the interaction between parameters and primarily suggested the optimal point for each interaction.

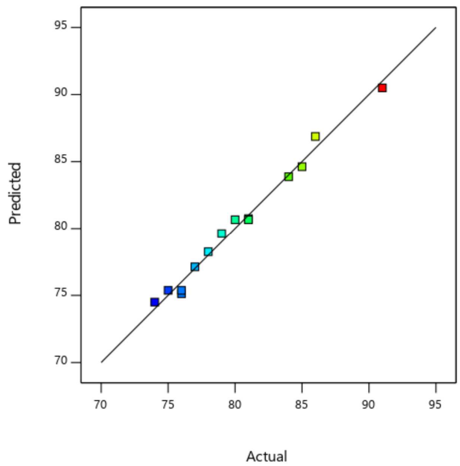

Figure 2. Actual and predicted leaching efficiency.

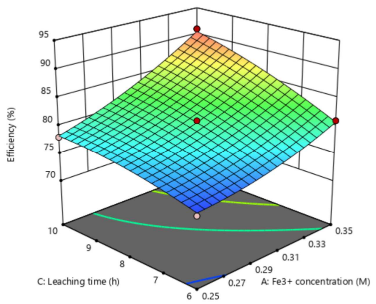

Figure 3. The response surface map of Fe^{3+} concentration vs. leaching time at H_2O_2 of 10 mL.

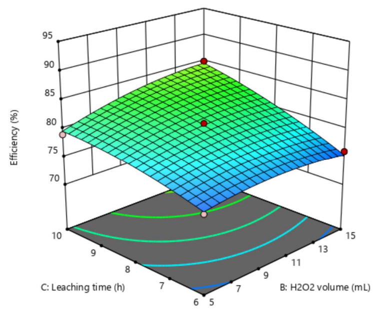

Figure 4. The response surface map of H_2O_2 volume vs. leaching time at Fe^{3+} of 0.3 M.

3.3. Optimum

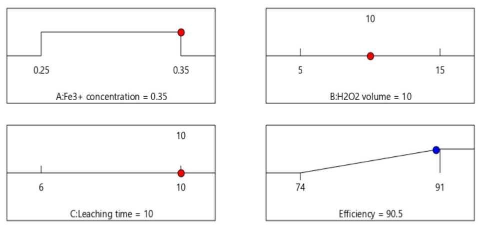

Figure 5. The optimal condition.

Design-Expert software was used to find the optimal condition. Further calculation showed that the optimal condition (Figure 5) with the highest efficiency of 90.5 % was achieved at Fe^{3+} concentration of 0.35 M, amount of H_2O_2 addition at 10 ml and leaching time of 10 h. In this study, leaching efficiency was higher than that from other references, such as the copper leaching efficiency of 85.86 % from refractory flotation tailings using H_2SO_4 [9], or the extraction copper of 83.8 %, by ethylenediaminetetraacetic acid (EDTA) from electronic waste [10]. So that, extraction by $Fe_2(SO_4)_3$ associated with the optimization process brings more efficiency. The results show that this research has improved than the others when optimizing the affecting factors by the response surface methodology to achieve high extraction efficiency. After leaching, copper was recovered from extract solution by electrochemical technology, resulted in the highest recovering efficiency of 85 percent.

4. CONCLUSION

The present study has successfully built the model, by response surface methodology, using Design Expert software, which is compatible with the experimental data, provided a high correlation coefficient (R^2). The optimal conditions were identified as $Fe_2(SO_4)_3$ concentration of 0.35 M, volume of H_2O_2 addition at 10 ml and leaching time of 10 h. Leaching efficiency of 90.5 percent and recovering efficiency of 85 percent was achieved. So that recycling copper in PCBs waste by the method proposed in this study is a promising technique for electronic waste treatment, which would minimize to the environment and human health effects, together with economic benefits.

REFERENCES

1. Perkins D. N., Brune Drisse M. N., Nxele T., Sly P. D. - E-Waste: A Global Hazard, Annals of Global Health **80** (2014) 286-295.
2. Gramatyka P., Nowosielski R., Sakiewicz P. - Recycling of waste electrical and electronic equipment, Journal of Achievements in Materials and Manufacturing Engineering **20** (2007) 535-538.
3. Sodhi M. S., Reimer B. - Models for recycling electronics end-of-life products, OR Spektrum **23** (2001) 97-115.
4. Sum E. Y. L. - The recovery of metals from electronic scrap, Journal of Metallurgy **43** (1991) 53-61.
5. Johan S., Shantha K. M. and Siti S. M. - A Review on Printed Circuit Board Recycling Technology, Journal of Emerging Trends in Engineering and Applied Sciences **3** (2012) 12-18.
6. Cui J. and Zhang L. - Metallurgical recovery of metals from electronic waste: A review, Journal of Hazardous Materials **158** (2008) 228-256.
7. Tan I. A. W., Ahmad A. L., and Hameed B. H. - Optimization of preparation conditions for activated carbons from coconut husk using response surface methodolog, Chemical Engineering Journal **137** (2008) 462-470.
8. Somasundaram M., Saravanathamizhan R., Basha C. A., Nandakumar V., Begum S. N., and Kannadasan T. - Recovery of copper from scrap printed circuit board: Modelling and optimization using response surface methodology, Powder technology **266** (2014) 1-6.
9. Xu B., Shuming W., Jian L. and Yilin L. - Response Surface Methodology for Optimization of Copper Leaching from Refractory Flotation Tailings, Minerals **8** (2018) 1-13.
10. Prashant J., Garima C., Pant K. K., Nigam K. D. P. - Greener approach for the extraction of copper metal from electronic waste, Waste Management **57** (2016) 102-112.