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Optimization of Recovery Process of Silver from Obsolete Printed Circuit Boards (PCBs)

Olusola Adesina[1*], Ajetunmobi Akeem[2] and Okewale Akindele[3]

[1]Department of Chemical and Petroleum Engineering, Afe-Babalola University, Ado-Ekiti, Nigeria.
[2]Department of Chemical Engineering, Obafemi Awolowo University, Ile-Ife, Nigeria.
[3]Department of Chemical Engineering, Federal University of Petroleum Resources, Effurun, Nigeria.

Authors' contributions

This work was carried out in collaboration between all authors. All authors wrote, read and approved the final manuscript.

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ABSTRACT

Aims: The aim of this work was to optimize the Silver recovery from Printed Circuit Boards (PCBs) of waste computers using statistical method by varying various parameters affecting the recovery.
Methodology: A total of 50 experimental runs was generated using central composite design, these were designed to study the effect viz; Thiourea conconcentration, H2SO4 conconcentration, Temperature, Particle diameter and time. Optimization was carried out using response surface methodology and validation of the result was also carried out.
Results: Statistically significant quadratic model was obtained. A quadratic model predicted the optimum silver recovery of 66.01% at optimal condition of thiourea concentration, 0.01 g/ml, sulphuric acid concentration, 0.01 M, particle size, 0.027 mm, temperature, 20°C and time of 60 min. The optimal condition was validated and 66.1% of silver recovery was obtained.
Conclusion: Response surface methodology was successively used to optimize thiourea leaching recovery of silver from PCBs of the waste computers. The result obtained could be scaled up to pilot plant.

**Corresponding author: E-mail: adesinaolusola50@yahoo.com;*

Keywords: Silver; printed circuit boards; response surface methodology; experimental design; optimization.

1. INTRODUCTION

The increasing in the volume of electronic waste (E- waste) has become a great concern to the world especially in a developing country such as Nigeria. Indiscriminate dumping of these materials has health implications on human, plant and animal. Due to the poor management of theses waste, many pollutants, such as persistent organic pollutants and heavy metals, are released from e-waste, which can easily accumulate into the food chain [1]. Owing to rapid developments in technology as well as tech-savvy customer's demand to accept new commodities, electronic products are being replaced by newer models at a much faster rate, predictions indicate that a huge numbers of e-waste will need to be disposed in the coming years [2]. Electronic waste components contain mercury, which can damage the brain; lead, which can damage reproductive systems; and cadmium, which causes kidney damage [3]. Also large parts of groundwater could also be polluted and render it unsafe for human consumption. E-waste is one of the fastest growing waste streams today and it is growing at three times the rate of municipal waste globally [4]. Information and communication technology (ICT) generated 53 million tons of e-waste as in 2012. Also 100 million phones are disposed of in Europe each year [5].

Developing country such Nigeria has become a dumping ground for e-waste, this is due to the importation of used electrical and electronic product to the country. Study showed that 176 containers of two categories of used electrical and electronic equipment were imported into Nigeria, from March to July 2010. About 500 shipping containers with 400,000 computer monitors or 175,000 large Television sets (TV) enter Nigeria each month [6]. One of the ways combating this is by recycling or reusing the components present in the waste. One of such useful components is silver.

Silver, is one of the widely used elements for industrial processing and in jewellery making. The majority of this element is consumed in film processing (40-50%), electrical and electronic industries (20-30%) and jewellery (10%) [7]. It is also important to note that there is an increase in demand for silver in the world, and one of the ways of meeting these demand is the recovery from the E-waste of electrical and electronic products. Although, silver is mostly recovered from scraps of X-ray films, photographic films and jewelry [8]. A certain amount can also be recovered from E-waste The most common leaching agents used in recovery of precious metals include cyanide, halide, thiourea, and thiosulfate [9,10,11]. Among all these agents only thiourea is able to achieve a highest recovery rate [12]. In acidic conditions, thiourea dissolves silver, forming a cationic complex. The reaction is rapid and silver/gold extractions of up to 99% can be achieved [12]. Some researches have been done on recovery of some precious metals from personal computers. Gurug et al. [13] worked on recovery of gold and silver from spent mobile phones using acidothiourea leaching. Jing-ying et al. [14] also studied the thiourea leaching of gold and silver from the printed circuit boards of waste mobile phones. However none of these authors optimized the process of recovery silver from the electronic components using response surface methodology (RSM).

Response-surface methodology comprises a body of methods for exploring for optimum operating conditions through experimental methods. Typically, this involves doing several experiments, using the results of one experiment to provide direction for what to do next. It can also be defined as experimental design and mathematical modelling, through the partial regression fitting of the experimental factors [15]. It has the advantage of reducing the number of experimental runs needed to give adequate information for statistically acceptable results. RSM model assesses the relationships between the response(s) and the independent variables [16], and defines the effect of the independent variables, alone or in combination, in processes [17].

In this present work recovery of silver from waste printed circuit boards of obsolete computers was investigated. In order to optimize the process, central composite design and RSM were applied to determine the effect of five factors (Thiourea conconcentration, Sulphuric conconcentration Temperature, Particle diameter and Time) on the silver recovery.

2. MATERIALS AND METHODS

2.1 Preparation of the Sample

The e-waste scrap was collected from the maintenance department of Obafemi Awolowo University Ile-Ife. Sorting of the e-waste samples into components was done by manual dismantling of the personal computers. The PCBs were then crushed using hammer milling, after which the particle size distribution of the crushed PCBs was done using an agitated sieve shaker of different mesh.

2.2 Leaching Process

Leaching of silver was carried out from crushed PCBs samples by using hydrometallurgical process of extracting metals through acidic thiourea solution because of its easiness to handle in the experiment design. In this process, crushed PCBs samples was contacted with thiourea and H2SO4 solution in a conical flask, and the heterogeneous mixture was shaken in a thermostated shaker at temperature as designed in the experiment for period of time to evaluate the effect of various parameters on the leaching reaction. The samples were then filtered and metal concentration in the filtrate was measured after proper dilution with distilled water [18]. Aliquots (10 ml) of the solution were withdrawn at appropriate intervals for determination of the amount of dissolved silver by volumetric analysis.

2.2.1 Silver content analysis

For the volumetric analysis, 10 ml of aliquot sample was taken in a conical flask and 5 ml of ferrous indicator was added. This solution was titrated against standard thiocyanate reagent to a permanent faint pink coloration [13]. The amount of silver in the sample was determined using the relation:

$$1ml\ of\ 0.1N\ KCNS \equiv 0.0108g\ Ag \qquad (1)$$

Where: N = Normal, KCNS = potassium thiocyanates and Ag = Silver.

At the end of each leaching process for a given contact time, the mixture was filtered using a Whatmann filter paper no 1 and the residue was then oven dried at 60°C for 6 hours after which it was reweighed. The percentage sample dissolution was done using (2).

$$\%\ sample\ dissolution = \frac{Weight\ difference\ of\ PCBs}{Weight\ of\ PCBs\ before\ leaching} X\ 100 \qquad (2)$$

2.3 Modeling and Optimization Studies of Silver Recovery

2.3.1 RSM experimental design

The Design-Expert 8.03 software was used to generate the experimental runs and modeling of the experimental data. Central composite design (CCD) was used to generate 50 experimental runs by considering five factors viz: Thiourea conconcentration (X_1), Sulphuric conconcentration (X_2), Temperature (X_3), Particle diameter (X_4), and Agitation time (X_5), using for each step of hydrolysis using BBD for each step of the hydrolysis. The coded independent variables levels are shown in Table 1.

Table 1. Coding of experiment factor and levels for silver recovery

Variable	Symbol	Coded levels		
		-1	0	+1
Thiourea conc (g/ ml)	X_1	0.01	0.05	0.09
Sulphuric acid conc (M)	X_2	0.01	0.05	0.09
Temperature (°C)	X_3	20	80	140
Time (min)	X_4	60	90	120
Particle size (mm)	X_5	0.2	0.4	0.6

2.3.1.1 Optimization

Response surface methodology (RSM) was used to optimize the quadratic model. The generalized response surface model for describing the variation in response variable is given below.

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i<j} \beta_{ij} X_i X_j + \varepsilon \qquad (3)$$

where Y is the predicted response by RSM, i and j are the linear and quadratic coefficients, respectively, β is the regression coefficient, k is the number of factors studied and optimized in the experiment, and ε represents the random error [19].

3. RESULTS AND DISCUSSION

Results of the analysis of variance (ANOVA) depicted in Table 3 showed F-value of 25.52, Since the corresponding P- value (*p* < 0.0001) for the model is less than 0.05, this implied that the model is significant and also showed that the model obtained is significant. The data obtained fitted well to a quadratic model and it exhibits low standard deviation [20]. It also had high coefficient of determination (R^2) of 0.95, this indicated the adequacy of the model for the representation of the actual relationship among the selected factors (Table 2) [21]. The p-values were used as a tool to check the significance of each of the coefficients, which in turn, are necessary to understand the pattern of the mutual interactions between the test variables [17]. Values of "Prob > F" less than 0.0500 indicated model terms were significant while values greater than 0.1000 indicated the model terms are not significant. If there are many insignificant model terms, they are not required to support hierarchy, in such situations and model reduction will improve the model. For the recovery of silver from PCB, p < 0.05 for each of the linear terms (X_1, X_2, X_3 X_4 X_5), cross-products (X_1X_2, X_1X_3, X_1X_3, X_1X_4, X_1X_5, X_2X_3, X_2X_4, X_4X_5) and quadratic terms (X_1^2, X_2^2 and X_3^2, X_5^2), these indicated that the model terms were significant. However quadratic terms X_4^2 and cross-products X_3X_5 were not significant and as such will not be included in the model equation. The final equation in terms of coded factors for the CCD response surface quadratic model is expressed in Y (reducing sugar concentration in g/l) in eq.4.

$$Y = 41.65 - 2.96X_1 - 1.36X_2 - 3.41X_3 - 2.37X_4 + 1.87X_5 + 1.58X_1X_2 + 2.62X_1X_3 + 3.11X_1X_4 - 2.34X_1X_5 + 5.1X_2X_3 + 5.06X_2X_4 + 1.39X_2X_5 + 3.61X_3X_4 + 3.77X_4X_5 - 3.61X_1^2 - 1.90X_2^2 + 1.63X_3^2 - 1.77X_5^2 \quad (4)$$

Figs. 1(a-j) depicts the various response surface plots for the recovery silver from PCBs. The curvature nature of the curves showed that there was an interaction between the response and the individual variables. It can also be deducted from the plot that low sulphuric acid concentration for the extraction contributed to increase in the recovery of the silver from PCBs, while lowest recovery was observed at highest concentration of the acid. The same trend was also observed for case of thiourea concentration. Increase in temperature and particle size favoured the recovery of silver from PCB, however high temperature led low recovery of the silver from the solution. The regression equation was solved by the design expert software and it predicted the optimum conditions for optimal recovery of silver. The optimal condition predicted was thiourea concentration of 0.01 g/ml, sulphuric acid concentration of 0.01 M, particle size of 0.027 mm temperature of 20°C and time of 60 min. The predicted recovery yield of silver was 66.01%. In order to verify the accuracy of the predictions by the software, the optimal condition values were applied to three independent replicates, by carrying out the optimal conditions experiments in the laboratory and the average yield obtained was 66.1% and this value is very close the one predicted by the software. This confirmed the efficacy of the mathematical model used [22].

Table 2. Central composite design (CCD) for silver recovery with five independent variables

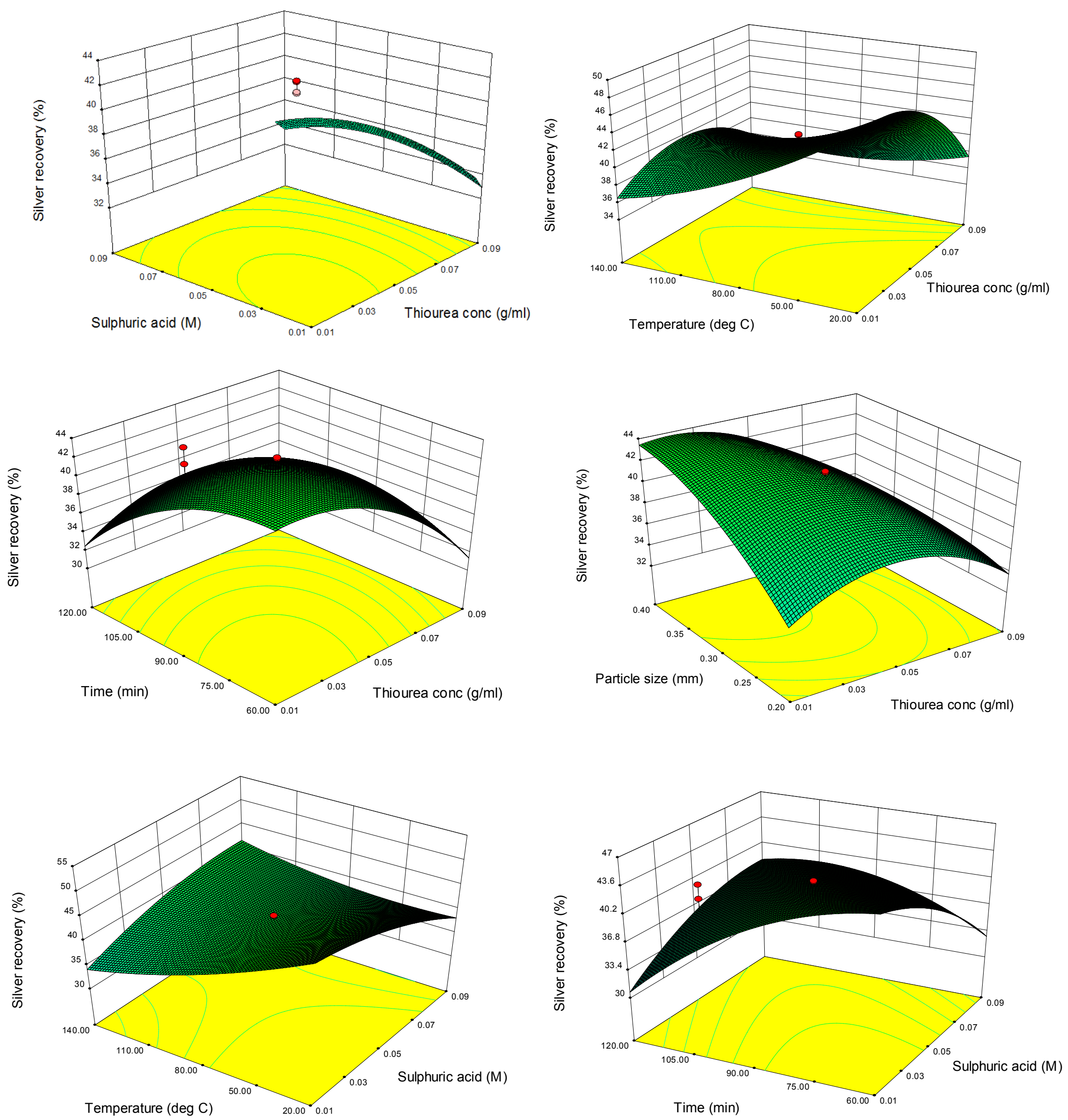
Run num	X_1	X_2	X_3	X_4	X_5	Experimental values	Predicted value
1	-1	-1	-1	-1	-1	33.01	32.90
2	1	-1	-1	-1	-1	33.90	36.16
3	-1	1	-1	-1	-1	39.01	36.11
4	1	1	-1	-1	-1	34.00	33.78
5	-1	-1	1	-1	1	44.00	44.67
6	1	-1	-1	-1	-1	33.62	35.72
7	-1	1	1	-1	-1	41.12	41.65
8	1	1	1	-1	-1	22.01	21.00
9	-1	-1	-1	1	-1	32.81	28.09
10	-1	1	-1	1	-1	41.12	41.65
11	1	1	-1	1	-1	19.70	20.55
12	-1	-1	1	1	-1	47.14	49.29
13	1	-1	1	1	-1	26.18	25.90
14	-1	1	1	1	-1	41.11	41.65
15	1	1	1	1	-1	38.07	38.10

Run num	X_1	X_2	X_3	X_4	X_5	Experimental values	Predicted value
16	-1	-1	-1	-1	1	14.00	14.33
17	1	-1	-1	-1	1	23.00	21.40
18	-1	1	-1	-1	1	41.22	41.65
19	1	1	-1	-1	1	41.12	41.65
20	-1	-1	1	-1	1	41.10	41.65
21	1	-1	1	-1	1	14.20	14.20
22	-1	1	1	-1	1	40.10	39.90
23	1	1	1	-1	1	42.41	48.68
24	-1	-1	-1	1	1	39.23	36.32
25	1	-1	1	1	1	29.01	27.45
25	-1	1	-1	1	1	41.20	42.54
27	1	1	-1	1	1	25.00	25.16
28	-1	-1	1	1	1	41.12	41.65
29	1	-1	1	1	1	25.02	27.15
30	-1	1	1	1	1	32.00	30.77
31	1	1	1	1	1	26.63	29.30
32	-1	0	1	1	1	24.60	23.46
33	1	0	0	0	0	25.00	26.62
34	0	-1	0	0	0	29.01	27.67
35	0	1	0	0	0	41.13	41.65
36	0	0	-1	0	0	28.20	25.98
37	0	0	1	0	0	21.02	19.05
38	0	0	1	0	0	24.30	27.20
39	0	0	0	1	0	29.90	30.60
40	0	0	0	1	0	37.40	36.32
41	0	0	0	0	1	29.20	30.36
42	0	0	0	0	0	31.02	29.78
43	0	0	0	0	0	58.00	63.31
44	0	0	0	0	0	40.02	39.36
45	0	0	0	0	0	76.70	65.13
46	0	0	0	0	0	21.02	22.35
47	0	0	0	0	0	42.12	41.65
48	0	0	0	0	0	32.80	34.14
49	0	0	0	0	0	40.00	39.36
50	0	0	0	0	0	42.01	41.65

Table 3. Analysis of variance (ANOVA) of regression equation

Source	Sum of Squares	df	Mean square	F-value	p-value
X_1	279.72	1	279.72	26.04	< 0.0001
X_2	80.38	1	80.38	7.48	0.0105
X_3	371.08	1	371.08	34.55	< 0.0001
X_4	199.32	1	199.32	18.56	0.0002
X_5	152.14	1	152.14	14.16	0.0008
$X_1 X_2$	79.54	1	79.54	7.40	0.0109
$X_1 X_3$	219.61	1	219.61	20.45	< 0.0001
$X_1 X_4$	309.57	1	309.57	28.82	< 0.0001
$X_1 X_5$	175.45	1	175.45	16.33	0.0004
$X_2 X_3$	832.42	1	832.42	77.50	< 0.0001
$X_2 X_4$	818.00	1	818.00	76.15	< 0.0001
$X_2 X_5$	61.86	1	61.86	5.76	0.0231
$X_3 X_4$	416.81	1	416.81	38.80	< 0.0001

Source	Sum of Squares	df	Mean square	F-value	p-value
$X_3 X_5$	0.029	1	0.029	0.0027	0.9586
$X_4 X_5$	453.98	1	453.98	42.27	< 0.0001
X_1^2	329.01	1	329.01	30.63	< 0.0001
X_2^2	195.59	1	195.59	18.21	0.0002
X_3^2	19.43	1	19.43	1.81	0.1891
X_4^2	228.92	1	228.92	21.31	< 0.0001
X_5^2	169.23	1	169.23	15.75	0.0004
Model	5483.30	20	274.17	25.52	< 0.0001
Residual	311.50	29	10.74		
Lack of Fit	308.41	19	16.23	52.55	
Pure Error	3.09	10	0.31		
Cor Total	5794.80	49			



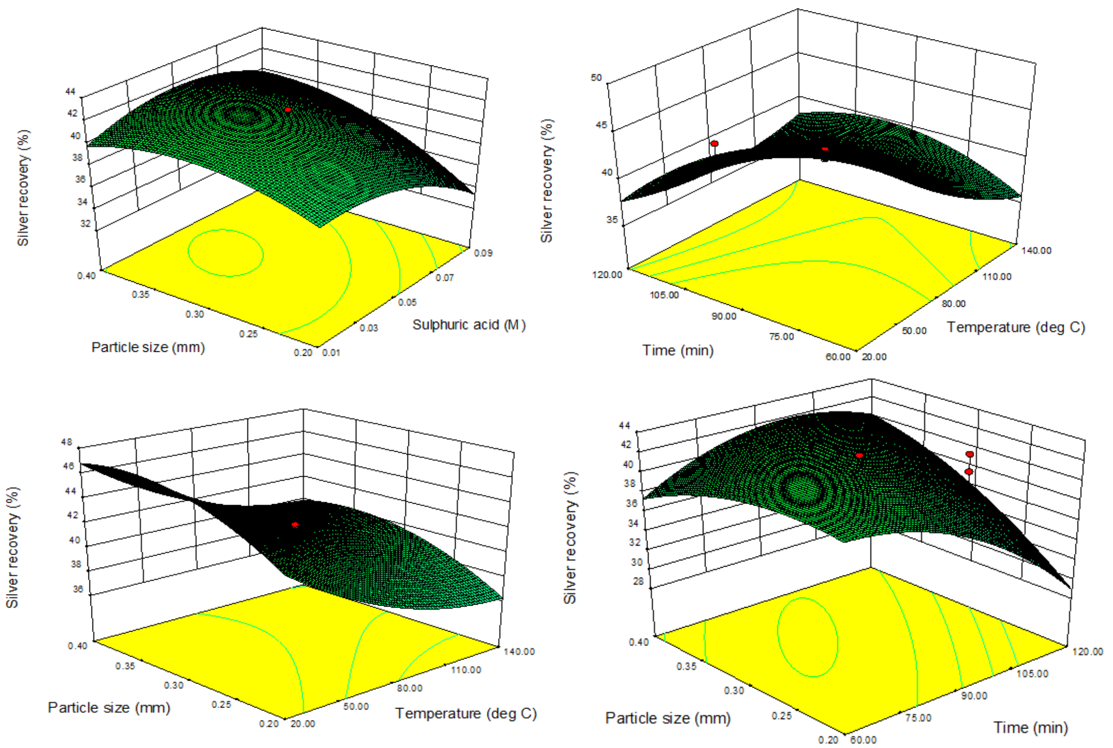


Fig. 1. Response surface plots for the recovery of silver from PCB

4. CONCLUSION

The work focused on statistical approach to the optimization of recovery of silver from printed circuit board (PCB). Effect of five independent factors: Thiourea concentration, H_2SO_4 concentration, Temperature, Particle diameter and leaching time on silver recovery was established. Analysis of variance (ANOVA) showed that all the factors have significant effect on the recovery of silver from PCB. A second-order mathematical model was obtained for the prediction of silver recovery, the optimal condition established was thiourea concentration, 0.01 g/ml, sulphuric acid concentration, 0.01 M, particle size, 0.027 mm, temperature, 20°C and leaching time of 60 min. The optimal condition was validated as 66.1 % of silver recovery. The work showed that silver could be recovered from printed circuit board and silver obtained can then be used for other industrial purposes

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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