Screening of variables affecting the selective leaching of valuable metals from waste mother boards PCBs

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Keywords: screening, leaching, metals, waste mother boards PCBs, Plackett-Burman Design

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Printed circuit boards (PCBs) are the main parts of waste electrical and electronic devices. Significant amounts of waste PCBs (roughly 1.5 million tons) are discarded yearly (Kaya, 2016). Waste mother boards PCBs obtained from obsolete computers can be considered as valuable secondary resources including base (such as Cu, Pb and Sn) and precious metals (such as Au and Ag). Moreover, most of these waste PCBs are incinerated or transferred to the landfill resulting in environmental and health problems owing to the production of toxic compounds such as furans and dioxins and leaching of some heavy metals such as lead into the landfill sites (Jadhav and Hocheng, 2015; Rocchetti *et al.*, 2013). Therefore, recycling the waste mother boards PCBs is required to recover valuable metallic resources and protect the environment at the same time.

With the advancement and development of technology in the manufacture of electronic equipment, the amount of precious metals in mother boards PCBs has considerably reduced. For this reason, recovery of base metals such as Cu, Pb, and Sn has received the most attention due to the presence of a significant concentration of these metals. In addition, the selective leaching of base metals in a preliminary step can lead to significantly improve subsequent recovery of precious metals. Lead which is used together with tin for soldering electronic components on the surface of mother boards PCBs also has a detrimental effect on human health and the environment when it encounters liquid waste streams created by industrial activities forming hazardous liquid solutions which can be absorbed by the soil and pollute the water (Parvez *et al.*, 2021).

Among the recovery processes of metals from waste PCBs including pyrometallurgy, hydrometallurgy and biometallurgy, hydrometallurgical process has attracted extensive attention due to its low environmental impact, low operational cost and ease of operation (Cui and Anderson, 2016). This process consists of pre-treatment of waste PCBs followed by leaching, adsorption and desorption stages. Leaching as the first stage of hydrometallurgical process using cyanide and acids such as nitric acid, hydrochloric acid and sulfuric acid is extensively applied owing to its simple process and low cost. However, the toxicity and corrosive problems of cyanide and acids limit the use of these lixiviants. Therefore, in this work, sodium thiosulfate and thiourea were used as lixiviants in an environmentally friendly manner to leach Cu, Pb, and Sn from waste mother boards PCBs.

Screening designs are generally used to reduce the number of experiments required for optimization of factors influencing on processes identifying the most significant parameters from a large number of suspected variables. A survey of literature proves that no studies have been carried out on the application of screening designs for the selective leaching of base metals from waste PCBs. In this study, after removing the inert coating from PCBs and crushing them to different particle size ranges, the PCB particles were treated using three strong acids in three consecutive stages to determine initial metal compositions using ICP-OES as can be seen in Table1: Stage1. Aqua regia (3 parts 35% HCl and 1 part 65% HNO_3 v/v) for 5 hours, stage 2. 35% HCl for 3 hours and stage 3. 65% HNO_3 during 3 hours at 80 °C and stirring rate of 500 rpm. Copper was found as the main component with significant quantities followed by tin and different amounts of other metals present in the PCBs. Then, Plackett-Burman Design as a powerful statistical tool was applied to screen the effect of six variables - leaching agent concentration (10-50 g.L^{-1})(A), temperature (20-60°C)(B), solid/liquid ratio (50-150 g.L^{-1})(C), initial pH (8-12 for sodium thiosulfate and 1-5 for thiourea which are stable in basic and acidic environments, respectively)(D), average particle size (0.3-1.5 mm)(E) and leaching time (4-10 h)(F) on the selective leaching of Cu, Pb and Sn in order to select the significant parameters. A six-factor 12-run Plackett-Burman screening design was generated using Statgraphics Version 18 as shown in Table 2. The experiments were performed at various combinations of high and low levels of the process variables to analyze their effect on the process assuming that there is no interaction between the parameters and is based on the first-order model:

$$Y_i = A_0 + \sum A_i X_i$$

Where Y_i are the response variables, A_0 is the scaling constant, A_i the regression coefficients of the response variables, and X_i are the independent variables.

The significant parameters were obtained based on 5% level (probability < 0.05) from the regression analysis. The results obtained for the recovery of Cu, Pb and Sn (in µg of metal/g of PCB) leached from waste mother boards PCBs showed that in general higher amounts of all the metals were leached using thiourea compared to sodium thiosulfate, and therefore, thiourea can be considered as a more efficient lixiviant to recover these base metals. The models obtained for Cu and Pb using thiourea were more significant with a higher coefficient of determination (R^2 = 0.821 and 0.795, respectively) whereas approximately the same R^2 was obtained for Sn using both lixiviants (0.809 for thiourea and 0.815 for thiosulfate). Initial pH as the most significant parameter was found to have a negative effect on Cu, Pb and Sn recovery using thiourea. In addition, thiourea concentration can be considered as a significant factor for all the metals since p values are very close to 0.05 (0.0728 for Cu, 0.0591 for Pb and 0.0650 for Sn) having a positive impact on Cu and negative on Pb and Sn leaching. Average particle size and solid/liquid ratio could be also considered to be significant on Sn leaching (p = 0.0756 and 0.0779, respectively) possessing negative impacts. In the case of sodium thiosulfate, an increase in thiosulfate concentration and temperature as the most significant parameters led to improve the recovery of Sn, whereas solid/liquid ratio was found to have a greater and negative impact on Cu recovery. No variables were found to be significant for Pb. The most significant parameters obtained from Plackett-Burman Design could be further optimized using Response Surface Methodology (RSM) models such as Central Composite Design (CCD) and Box-Behnken Design (BBD).

Table 1. Initial metal compositions of waste mother boards PCBs (wt %).

Metal / Particle Size,	Na	Mg	Al	K	Ca	Fe	Ni	Cu	Zn	Sn	Pb	Sb	Pd	Ag	Au
0.1-0.5	1.409	0.188	1.723	0.053	3.713	0.091	0.085	37.942	0.462	4.144	1.163	0.006	0.0005	0.056	0.002
0.5-1.0	2.095	0.231	1.325	0.069	3.359	0.073	0.295	57.167	5.127	3.885	1.009	0.006	0.0003	0.068	0.003
1.0-2.0	1.661	0.208	1.698	0.056	4.299	1.002	0.101	54.652	1.167	6.247	0.296	0.009	0.0002	0.058	0.001

Table 2. Plackett-Burman experimental designs for leaching of Cu, Pb and Sn using thiourea and sodium thiosulfate.

Run	A	B	C	D	E	F	Thiourea			Sodium Thiosulfate		
							Cu Conc. (µg/g)	Pb Conc. (µg/g)	Sn Conc. (µg/g)	Cu Conc. (µg/g)	Pb Conc. (µg/g)	Sn Conc. (µg/g)
1	10	20	150	5[a] / 12[b]	1.5	4	676.963	0.0095	0.4362	211.771	0.0044	0.3345
2	10	60	150	1 / 8	1.5	4	12638.7	590.22	69.904	4583.68	0.0470	0.0385
3	10	20	50	5 / 12	1.5	10	1428.87	0.0259	0.3839	269.011	0.0000	0.1884
4	50	20	150	5 / 12	0.3	10	2830.10	0.0106	0.6979	3749.13	0.2800	1.8467
5	50	20	50	1 / 8	1.5	10	106962	66.949	0.7661	14192.3	0.0000	2.3931
6	10	60	150	5 / 12	0.3	10	1561.53	0.3020	0.0222	66.4960	11.838	0.5739
7	10	20	50	1 / 8	0.3	4	22391.5	2039.5	1164.7	2551.85	0.0000	0.0919
8	10	60	50	1 / 8	0.3	10	28506.7	1175.0	1045.2	20543.4	0.4029	0.0430
9	50	60	50	5 / 12	0.3	4	14055.4	0.0827	0.2884	11382.0	1.7183	26.675
10	50	60	50	5 / 12	1.5	4	13057.6	0.2659	0.1359	11816.3	1.9720	76.004
11	50	60	150	1 / 8	1.5	10	63996.1	70.689	3.5361	7117.25	0.0749	23.773
12	50	20	150	1 / 8	0.3	4	20934.6	7.1865	12.034	4062.39	0.2578	2.4497

a) Initial pH for thiourea b) Initial pH for sodium thiosulfate

Acknowledgements

This work is part of I+D+i project Reference PID2021-122923NB-I00 financed by MCIN/AEI /10.13039/501100011033 / FEDER, UE. Authors would like to thank the use of RIAIDT-USC analytical facilities.

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