



# Optimization of metal bio-acid leaching from mobile phone printed circuit boards using natural organic acids and H<sub>2</sub>O<sub>2</sub>

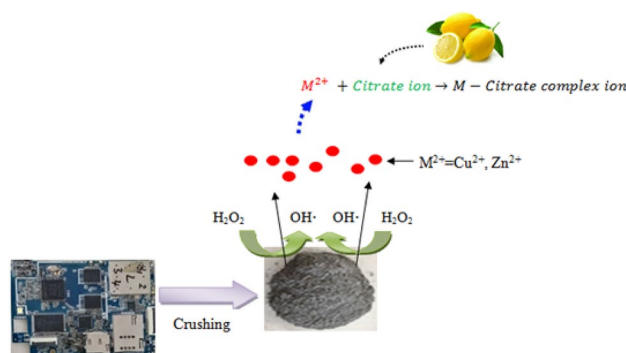
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## Abstract

With increased popularity and technological innovation, more and more electric and electronic equipment wastes (e-wastes) are being generated. Printed circuit boards (PCBs) as fundamental components of electronic wastes, consist of both precious and heavy metals. In this research, the simultaneous extraction Cu and Zn from PCBs used in mobile phones with a natural organic acid and H<sub>2</sub>O<sub>2</sub> was examined. The organic acid was determined from agricultural lemon fruit. The leaching extraction factors investigated include lemon juice concentration, solid/liquid (S/L) ratio, and H<sub>2</sub>O<sub>2</sub> concentration. Response surface methodology (RSM) was applied to optimize the extraction which resulted in the maximum recoveries of 89% Cu and 73% Zn at 1.41% (w/v) S/L ratio, 12.2% (v/v) H<sub>2</sub>O<sub>2</sub>, and 74% (v/v) lemon juice after 4 h at 20 °C. The effect of time on the Cu and Zn recoveries at the determined optimum condition was examined. Furthermore, a kinetic study based on a shrinking core model was performed and the result showed that mass diffusion was rate-limiting in the extraction.

## Graphic abstract



**Keywords** Natural organic acid · Printed circuit boards · Extraction · Optimization · Kinetics

## Abbreviations

ANOVA Analysis of variance  
C.I. Confidence interval  
CCD Central composite design  
E-wastes Electronic wastes

FE-SEM Field emission scanning electron microscopes  
HPLC High-performance liquid chromatography  
ICP-OES Inductively coupled plasma optical emission spectrometer  
PCBs Printed circuit boards  
RSM Response surface methodology  
S/L ratio Solid/liquid ratio  
S/N Signal to noise

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## Introduction

The short lifespan of electronics and their increased popularity in the consumer market have turned the issue of obsolete electronics a big problem in the modern world [1]. Electronic waste is produced at a faster pace than other waste nowadays [2]. Each year, 20–50 million metric tons of electronic waste is produced every year worldwide [3, 4]. E-waste recycling is an effective tool for e-waste management that benefits the environment [5]. Electronic equipment consists of heavy metals such as Pd, Hg, As, Cd, Se, and Cr, which can cause serious health problems [6, 7]. E-waste is also known as artificial mines because they contain large quantities of valuable metals including Au, Ag, and Cu. A ton of mobile phones contains 3.5 kg Ag, 340 g Au, 130 kg Cu, and 140 g Pd [8]. The amount of Cu in computer PCBs was estimated to be up to 400 kg, larger than 250 kg Cu in a Cu mine [9, 10].

The metals recycling from e-waste is important in terms of both waste management and economics. Recycling technologies for waste management should be simple, low-cost, and do not produce new wastes [11]. There are studies which used traditional methods including pyrometallurgy and hydrometallurgy to extract precious metals from PCBs. Mizero et al. [12] examined the extractions of Cu, Ni, and Zn from two samples of mobile phone and computer PCBs using sulfuric acid and  $H_2O_2$ . Their results showed 70% and 80% of Cu extraction for batch reactor and agitator reactor, respectively, with 4.8 M  $H_2O_2$ , 4.3 M sulfuric acid, 0.5 mm waste particle size, 194 rpm mixing rate and 0.1% S/L. Jha et al. [13] investigated the extraction of Pb from the soldering materials on PCBs using nitric acid. They determined nearly 100% Pb extraction with 0.2 M nitric acid at 90 °C and 0.1% S/L after 2 h. Using this traditional method consumes a large amount of energy and are less efficient [14]. It is obvious that hydrometallurgical methods require high concentrations of acids, which causes an environmental hazard [15, 16].

Bioleaching which relies on acids secreted by microbes is a new, effective, economical and environmentally friendly way to extract metal elements from e-waste [17, 18]. Various bacteria and fungi are utilized in bioleaching. The most commonly used microbes include *Acidithiobacillus ferrooxidans*, *Acidithiobacillus thiooxidans*, and *Aspergillus niger* [19, 20]. The bioleaching of obsolete PCBs has been reported in the literature [9, 21, 22]. Arshadi and Mousavi [22] studied bioleaching of Cu and Au from PCBs using a pure culture *Bacillus megatherium* and also a pure culture of *Cyanogenic bacterium*. Maximum Cu recovery in the sample without adjusting pH was 69% and 100% with adjustment pH = 10. In another study, Arshadi and Mousavi [9] used RSM to optimize

Cu and Ni extractions from PCBs using *A. ferrooxidans*. Approximately 100% of Cu and Ni were extracted under optimum conditions including an initial pH of 1, initial  $Fe^{3+}$  concentration of 4.18 g/L, pulp density of 8.5 g/L and particle size of 114.02  $\mu m$ . Although bioleaching is safe and environmentally friendly, it needs a long time and also different salts in the culture media to culture microbes which are costly [15, 20].

Therefore, there is a need for a new method which not only has environmentally friendliness like bioleaching but also needs fast processing time. This new way uses the biogenic acid obtained from natural organic sources such as lime or lemon that are rich in organic acids such as citric acid, maleic acid, and ascorbic acid. Using these bio-acids not only are environmentally friendly but also are good at resolving slow kinetics of bioleaching [15, 23]. Rahimi et al. [15] used natural bio-acids from lemon juice to recover of vanadium from fly ash which achieved nearly 100% recovery after 3 h. In another study, Esmaeili et al. [23] used lemon juice to extract valuable metal from lithium ion batteries (LIBs) with recovery yields of 100% Li, 96% Co, and 96% Ni after 45 min. In the literature, no studies have been reported using natural organic acids to extract precious metals from PCBs. Therefore, in this work, Cu and Zn extractions from PCBs were studied using natural organic acids from lemon juice. Different process operating factors including lemon juice concentration,  $H_2O_2$  concentration, and S/L ratio were optimized using RSM. Furthermore, the kinetics of the bio-acid leaching process was performed to determine the rate-limiting step of the overall reaction.

## Materials and methods

### Preparation of PCBs

Samples of PCBs were collected from a local electrical equipment repair center in Sanandaj, Iran. The PCBs were grounded and sieved with mesh size 149–177  $\mu m$ . To determine the amounts of Cu and Zn in a sample, 0.5 g particles were dissolved in 30 mL aqua regia and put on a magnetic heater for 24 h. An inductively coupled plasma optical emission spectrometer (ICP-OES) (Vista Pro, Varian, Inc., California, USA) was used to determine the metal contents which were (mg/L) Pb 34.54, Ni 142.2, Fe 172.7, Ag 20.2, Cu 497, and Zn 37.26.

### Lemon juice preparation

Fresh lemons were procured locally in Sanandaj, Iran. They were squeezed by hand. The lemon juice was centrifuged to separate any pulp and debris. The amounts of organic acids in the lemon juice were determined using a

high-performance liquid chromatography (HPLC) (Model LC20AD, Shimadzu, Japan).

### The design of experiments

RSM is a set of mathematical methods to determine the relationships between one or more variable responses with multiple independent variables. RSM based on central composite design (CCD) with three factors in five levels with six center points was used to optimize Cu and Zn extractions. The analysis of variance (ANOVA) was used to determine model variables. The effects of three factors of lemon juice concentration, S/L ratio, and H<sub>2</sub>O<sub>2</sub> concentration on Cu and Zn recoveries were investigated. The levels of each variables and the experiment list based on CCD resulted in 20 runs shows in Table 1.

### Bio-acid leaching experiment

All experiments were carried out in 100 mL beakers stirred magnetically at 200 rpm at 20 °C for 4 h. The samples were filtered with filter paper and then the filtrate is centrifuged at 3000 rpm for 15 min at the end of each experiment and the concentrations of Cu and Zn ions were determined using ICP-OES. More detailed explanations about the testing procedures and also some photographs of experiments come in

**Table 1** Experimental design based on CCD

Run	S/L ratio % (w/v)	H <sub>2</sub> O <sub>2</sub> % (v/v)	Lemon juice % (v/v)	Recovery (%)	
				Zn	Cu
1	2.7	8.0	70.0	49.5	42.2
2	1.4	3.8	87.8	38.5	23.5
3	2.7	8.0	100	55.3	44.1
4	2.7	8.0	70.0	56	46.4
5	0.5	8.0	70.0	69.3	69.6
6	4.1	12.2	52.2	56.1	35.2
7	1.4	12.2	87.8	72	86.2
8	2.7	8.0	70.0	47.4	44.1
9	2.7	8.0	70.0	57.6	47.1
10	2.7	15.0	70.0	60.2	55.9
11	1.4	12.2	52.2	74.6	80.9
12	2.7	8.0	70.0	40.3	45.0
13	1.4	3.8	52.2	42.3	34.6
14	5	8.0	70.0	38.7	25.5
15	2.7	1.0	70.0	18.4	3.7
16	4.1	3.8	87.8	20.8	3.1
17	2.7	8.0	70.0	55.3	39.6
18	2.7	8.0	40.0	53.6	32.2
19	4.1	12.2	87.8	41.9	27.4
20	4.1	3.8	52.2	31.1	13.6

Fig. 1. Metal extraction recovery was determined from the following equation [15]:

$$R(\%) = \frac{C_1 V_1}{C_2 V_2} \times \frac{M_2}{M_1} \times 100, \tag{1}$$

where *R* is the Cu and Zn leaching efficiencies (%), *C*<sub>1</sub>, *M*<sub>1</sub>, and *V*<sub>1</sub> are the metal concentrations (mg/L), solid mass and liquor volume in the bio-acid leaching solution, respectively. *C*<sub>2</sub>, *M*<sub>2</sub>, and *V*<sub>2</sub> are metal concentrations, solid mass, and aqua regia volume, respectively, in aqua regia leaching. Additionally, surface characteristics of the particles from crushed PCBs before and after the bio-acid leaching were analyzed under a field emission scanning electron microscope (FE-SEM) (TESCAN, Czech Republic).

## Results and discussion

### Statistical analysis

The required experiments were performed in accordance with RSM with face CCD that needed 20 runs. Zn and Cu recoveries were selected as responses (Table 1). The percentages of Cu and Zn extractions from the RSM method were found to be:

$$\begin{aligned} \text{Cu recovery (\%)} = & + 44.06 - 13.20A + 15.53B + 3.54C \\ & - 7.91AB - 1.54AC + 2.36BC \\ & + 1.26A^2 - 5.07B^2 - 2.12C^2 \\ & - 1.74ABC + 3.81A^2B - 6.52A^2C - 5.01AB^2, \end{aligned} \tag{2}$$

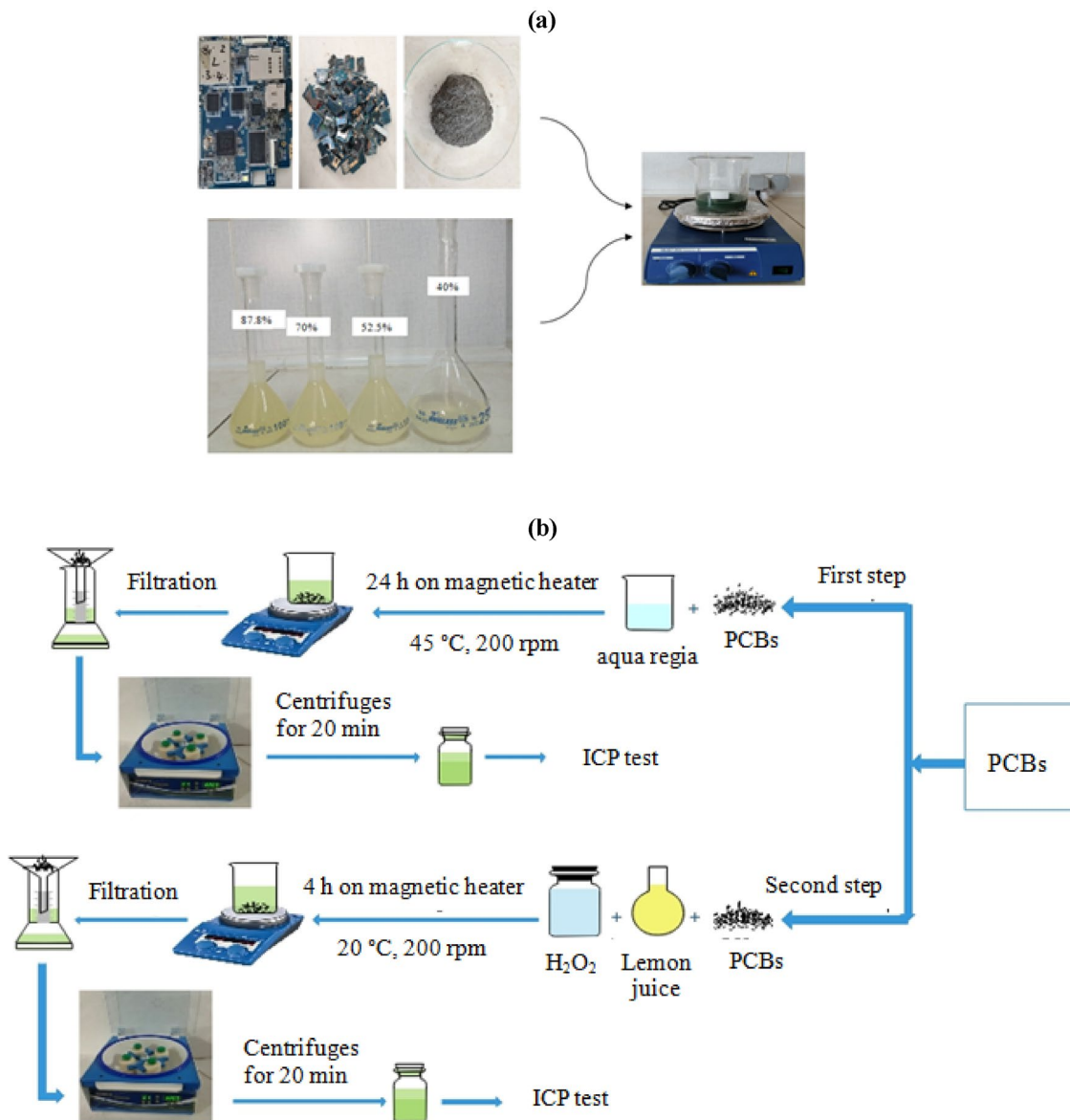
$$\begin{aligned} \text{Zn recovery (\%)} = & + 52.15 - 9.44A + 13.34B - 2.05C \\ & - 2.46AB - 2.26AC - 0.34BC - 4.69B^2 \end{aligned} \tag{3}$$

In Eqs. 2 and 3, A, B, and C are encoding parameters for S/L ratio, H<sub>2</sub>O<sub>2</sub> concentration, and lemon juice concentration, respectively. The results of ANOVA are shown in Table 2. Based on the ANOVA analysis, the reduced cubic model for Cu and the reduced quadratic model for Zn were statistically significant with *p* value < 0.0001, which show that the determined models are significant for studying optimal conditions and the high level of reliability. The adequate precision, reflecting the signal to noise (S/N) ratio, was 38.9 for Cu and 17.3 for Zn, much larger than the common threshold of 4.

### Contour plots

#### Cu recovery

Figure 2a shows the effects of H<sub>2</sub>O<sub>2</sub> concentration and S/L ratio with fixed 70% (v/v) lemon juice on Cu recovery. The

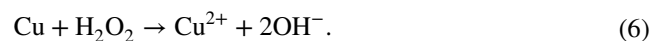


**Fig. 1** **a** Some photographs of the experiments; **b** the schematic of the experimental procedure

results showed that increasing  $\text{H}_2\text{O}_2$  concentration from 3.84 to 12.2% (v/v) and decreasing S/L ratio from 4.09 to 1.41% (w/v) resulted in increased Cu recovery from 16.2 to 76.0%. The positive effect of  $\text{H}_2\text{O}_2$  was attributed to increased solution potential ( $E_h$ ) that led to the breaking of the chemical bond of Cu from solid matrices [23]. According to Purbe diagram in Hong et al. [24], Cu can change to ion form at high solution  $E_h$  0.34 V, which was achieved by adding  $\text{H}_2\text{O}_2$ . Equations 4–6 show how  $\text{H}_2\text{O}_2$  is separated and reacted with Cu. According to these equations, by decomposing hydrogen peroxide and producing hydroxyl radicals, the oxidation potential of the environment increases and as a result, copper enters the solution phase as an ion [24].



Equations 4 and 5 show the anodic reaction of Cu dissolution and cathodic reaction of  $\text{H}_2\text{O}_2$  reduction, respectively. The overall redox reaction of Cu in  $\text{H}_2\text{O}_2$  solution is shown below [24]:



According to Fig. 2a, effect of  $\text{H}_2\text{O}_2$  on Cu recovery is depend on the amount of S/L ratio. It is determined the  $\text{H}_2\text{O}_2$

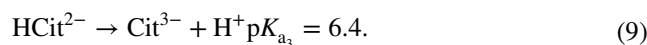
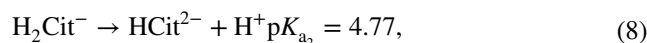
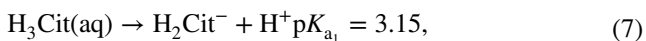
**Table 2** ANOVA analysis

Source	S.S.	df	M.S.	F value	p value	
Model Cu recovery	9186	13	707	109	<0.0001	Significant
A	986	1	986	152	<0.0001	
B	1363	1	1363	2190	<0.0001	
C	70.8	1	70.8	10.9	<0.0001	
AB	501	1	501	77.1	<0.0001	
AC	18.9	1	18.9	2.90	0.14	
BC	44.6	1	44.6	6.83	0.040	
A <sup>2</sup>	22.9	1	22.9	3.52	0.11	
B <sup>2</sup>	371	1	371	57.0	0.0003	
C <sup>2</sup>	64.5	1	64.5	9.92	0.020	
ABC	24.2	1	24.2	3.72	0.10	
A <sup>2</sup> B	48.1	1	48.1	7.40	0.035	
A <sup>2</sup> C	141	1	141	21.7	0.0035	
AB <sup>2</sup>	83.2	1	83.2	12.8	0.012	
Residual	39.0	6	6.50			
Lack of fit	0.049	1	0.049	6.25	0.94	Not significant
Model Zn recovery	4119	7	588	22.2	<0.0001	Significant
A	1218	1	1218	45.9	<0.0001	
B	2430	1	2431	91.6	<0.0001	
C	57.6	1	57.6	2.17	<0.0001	
AB	48.5	1	48.5	1.83	0.201	
AC	41.0	1	41.0	1.54	0.238	
BC	0.91	1	0.91	0.034	0.856	
B <sup>2</sup>	323	1	323	12.2	0.0045	
Residual	319	12	26.6			
Lack of fit	102	7	14.6	0.34	0.906	Not significant

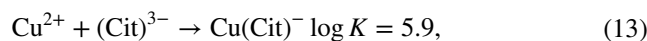
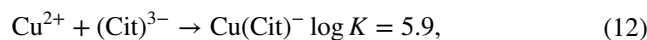
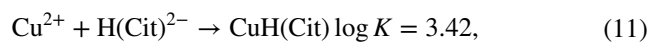
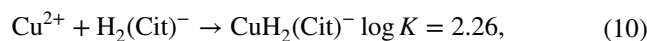
\*Cu recovery:  $R^2=0.99$ ;  $\text{adg.}R^2=0.98$ ; C.V. (%) = 6.37; Adeq. precision = 38.9; Zn recovery:  $R^2=0.92$ ;  $\text{adg.}R^2=0.88$ ; C.V. (%) = 10.5; Adeq. precision = 17.3

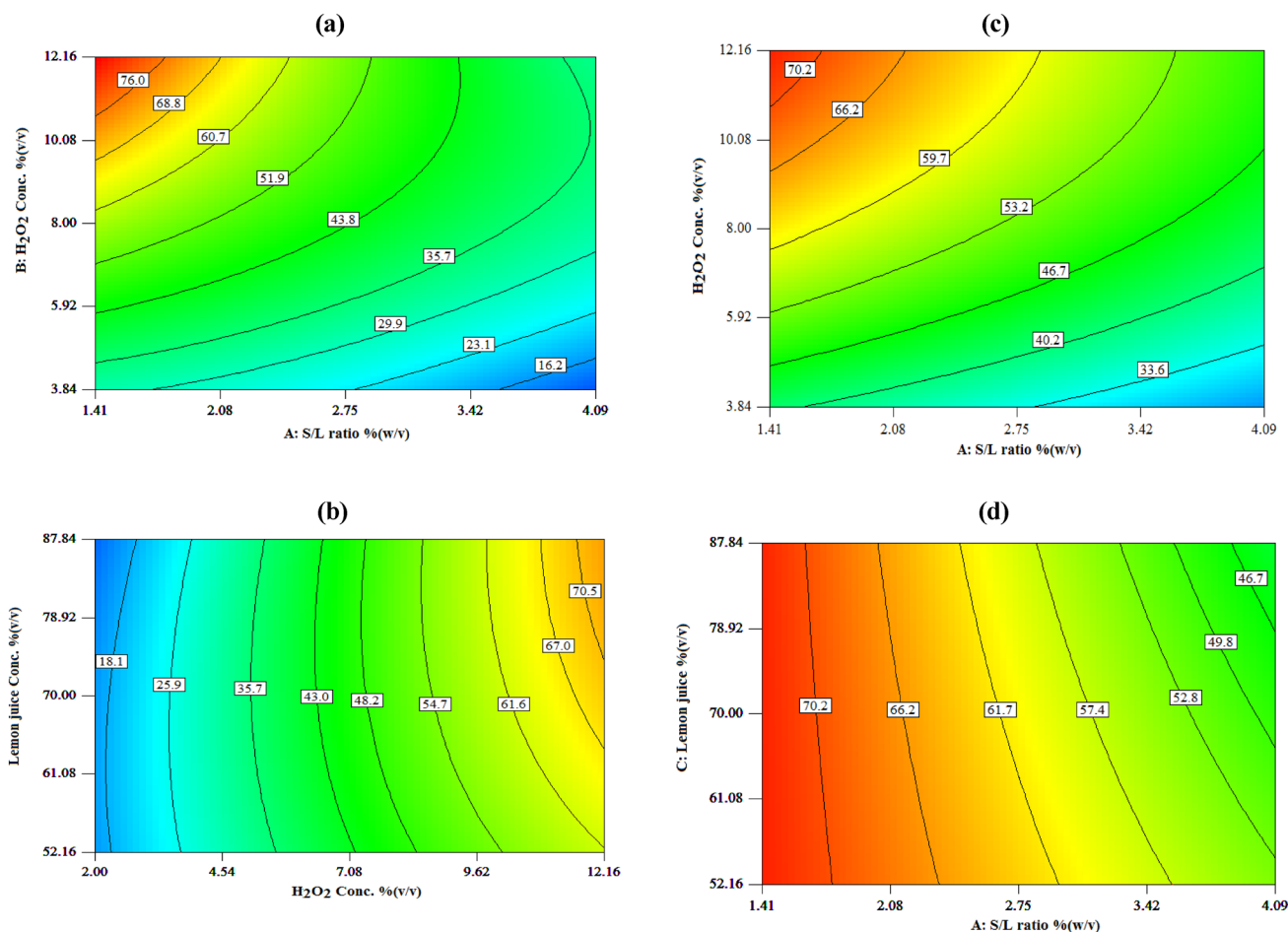
has lower effect at high amount of S/L ratio. Increasing  $\text{H}_2\text{O}_2$  from 3.84 to 12.16% (v/v) at S/L ratio 1.41% (w/v) cause increasing Cu recovery from 29.9 to 76.0% (improve 66.1% efficiency) but at S/L ratio 4.09% (w/v) cause increasing Cu recovery from 16.22 to 35.7% (improve 19.5% efficiency). This negative effect of increasing S/L ratio on Cu extraction is due to the high viscosity of the mixture and, therefore, increased mass transfer resistance.

Figure 2b shows the interaction between lemon juice and  $\text{H}_2\text{O}_2$  on Cu recovery at S/L ratio 1.41% (w/v). It indicates that the maximum 70.5% Cu recovery can be determined at 12.2% (v/v) of  $\text{H}_2\text{O}_2$  and 87.8% (v/v) lemon juice. Increasing lemon juices increased bio-acids like citric acid, ascorbic acid, and maleic acid concentration. This resulted in a lower pH and the metals in the waste were extracted more efficiently. Equations (7)–(9) show the hydrogen ion ( $\text{H}^+$ ) production from citric acid [15]:



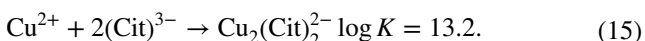
Additionally, the presence of different forms of protonated citric acid species in the solution (including  $\text{H}_2\text{Cit}^-$ ,  $\text{HCit}^{2-}$ , and  $\text{Cit}^{3-}$ ) acted as chelating agents forming metal complexes as shown below which can increase the leaching efficiency [24]:





**Fig. 2** Interactions between different factors: **a** effects of H<sub>2</sub>O<sub>2</sub> concentration and S/L ratio parameters on Cu recovery, and **b** effects of H<sub>2</sub>O<sub>2</sub> concentration and Lemon juice concentration on Cu recovery; **c**

effects of H<sub>2</sub>O<sub>2</sub> concentration and S/L ratio parameters, and **d** effects of H<sub>2</sub>O<sub>2</sub> concentration and Lemon juice concentration on Zn recovery



Based on pH–Eh diagram only cupric ions (Cu<sup>2+</sup>) can complex with citric acid and the formation of complexes are different as a function of solution pH. These Cu ions are easily complexed by citrate ions dissociated from the citric acid, which accelerated the leaching rate of Cu. As the pH of 4 (which is the pH of lemon juice), dissolved Cu<sup>2+</sup> ions can form the complex with citrate (H<sub>2</sub>Cit<sup>-</sup>) ions [24].

**Zn recovery**

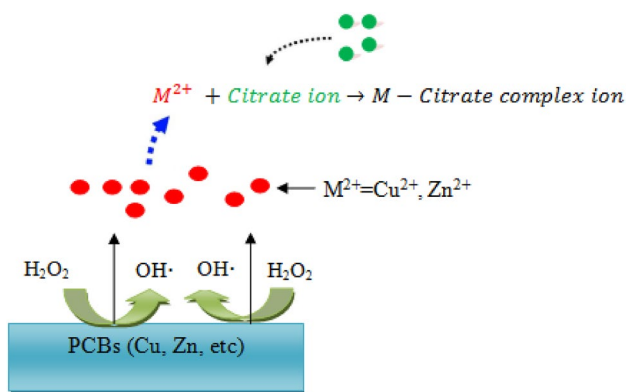
The interactions and effects of H<sub>2</sub>O<sub>2</sub> concentration at different S/L ratios for Zn extraction with 70% (v/v) lemon juice are shown in Fig. 2c. The results indicated that Zn recovery increased from 42.3 to 70.4% for H<sub>2</sub>O<sub>2</sub> concentration from

3.84 to 12.16% (v/v). Furthermore, the increase in S/L ratio had a negative effect on Zn extraction. At H<sub>2</sub>O<sub>2</sub> concentration of 3.84% (v/v), increasing the S/L ratio from 1.41 to 4.09% (w/v) decreased the extraction efficiency from 40.2 to 33.6%. By reducing the residual density and viscosity of the liquor, solid particles were suspended evenly in the leaching solution, so the leaching agent penetrated the internal solid particles more easily and thus increased the chances for encounters between them [25].

Figure 2d also shows the effects of lemon juice and S/L ratio on Zn recovery at 11.8% (v/v) of H<sub>2</sub>O<sub>2</sub>. The maximum recovery of Zn was 70.2% at 1.41% (w/v) S/L ratio, and 87.8% (v/v) of lemon juice.

**The mechanism of bio-acid leaching with H<sub>2</sub>O<sub>2</sub>**

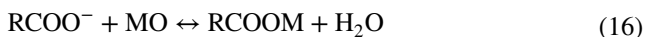
Based on Fig. 2, it is obvious to achieve high recovery efficiencies of both Cu and Zn, high concentrations H<sub>2</sub>O<sub>2</sub> and lemon juice were needed. Figure 3 shows the mechanism of



**Fig. 3** The mechanisms for H<sub>2</sub>O<sub>2</sub> and citrate to enhance metal recovery from PCBs

enhancement of metal recovery in the presence of H<sub>2</sub>O<sub>2</sub> and citric acid as their highest concentrations. H<sub>2</sub>O<sub>2</sub> attacked PCBs first and extracted M<sup>2+</sup> (Cu<sup>2+</sup> and Zn<sup>2+</sup>) from the particles to the liquor. Then citrate ion reacted with M<sup>2+</sup> to form a complex. Based on Le Chatelier’s principle, citrate consumed M<sup>2+</sup> in the medium and resulted in enhanced metal extraction using H<sub>2</sub>O<sub>2</sub>.

Steer and Griffiths [26] noted that Zn extraction might be better explained by substituent group effects from Lewis acid/base theory. The substituent groups attached to the carboxylic acid functionality can play an important role. Although carboxylic acids are only partially dissociated “weak acids”, the carboxylate anion (RCOO<sup>−</sup>) shown in Eq. 16 could potentially alter the extraction capability.



M—metal; R—organic substituent group.

**Optimization and model confirmation experiment**

The main aim of process optimization was to receive the maximum Cu and Zn extractions. At the optimum conditions, the extraction yields were 89% for Cu and 73% for Zn. The optimum conditions were 74% lemon juice, 12.2% H<sub>2</sub>O<sub>2</sub> and 1.41% S/L ratio. To validate the model, an experiment using the optimum conditions predicted by the model was performed. Table 3 results indicate that Cu and Zn extraction yields were in the range ±95% confidence interval (C.I.) which it can be concluded that the model was verified to the predicted experimental condition. Comparing the optimum

result of this study with previous researches has been done (Table 4). The result shows the present research using bio-acid leaching method based on lemon juice has been able to extract metals with much less dependence on pH, temperature, residual toxicity, higher extraction efficiency in a shorter time than the bioleaching methods. Also, using bio-acid compared to chemical acid leaching studies, due to the use of natural acid in the lemon juice does not cause environmental pollution and it is much more economical.

**HPLC and SEM analysis**

The acid content in lemon juice was measured using HPLC. Result found 90 mg/g citric acid, 0.86 mg/g malic acid, and 1.24 mg/g ascorbic acid. The present of these form organic acid in the lemon juice could increase metal leaching from solid matrix. To show the effects of bio-acid extraction, field emission scanning electron microscopes (FE-SEM) were used to analyze the particle surface of the waste sample before the leaching process (Fig. 4a) and the after the leaching process (Fig. 4b). A comparison of the images before and after the leaching process shows that lemon juice was able to erode the particle surface considerably, making metal extraction possible.

**Kinetics study**

After determining the optimal condition using RSM, the effects of Cu and Zn recovery yields at these conditions were examined for different times up to 5 h and the results are shown in Fig. 5. It is obvious that the recovery yields of Cu and Zn were almost constant at 150 min, but the highest recovery yields reached 90% Cu and 74% Zn after 300 min. A shrinking core model was employed. In this model, solid particles are treated as having spherical and non-porous, which are surrounded by a thin layer of leaching solution. Mass transfer takes place between solid and fluid film. According to the model, if the diffusion is the rate-limiting step, the model is shown in Eq. 17, and if the chemical reaction is the rate-limiting step, the model is shown in Eq. 18.

$$kt = 1 - 3(1 - X)^{\frac{2}{3}} + 2(1 - X), \tag{17}$$

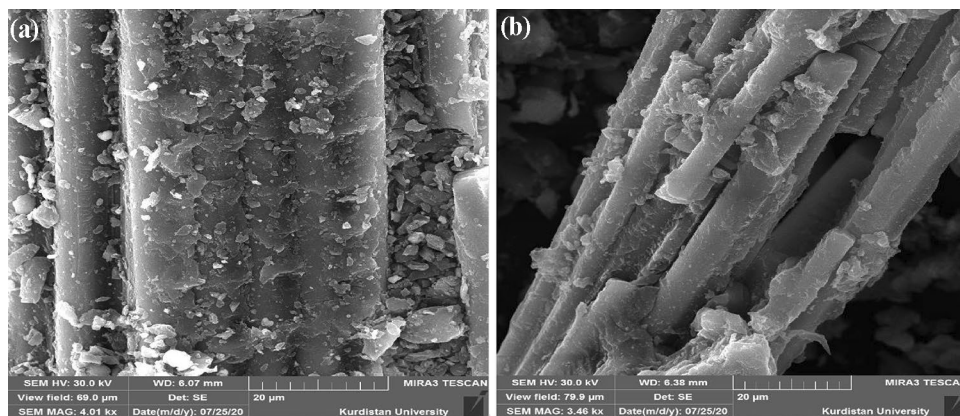
$$kt = 1 - (1 - X)^{\frac{1}{3}}, \tag{18}$$

**Table 3** Confirmation of the predicted optimal conditions

Response	Goal	Predicted by the model	Experimental result	95% C.I. low	95% C.I. high
Cu recovery (%)	Maximize	86.2	89	81.8	90.6
Zn recovery (%)	Maximize	72.7	73	66.1	79.3

**Table 4** Comparison of research on the extraction of precious metals from PCBs

Method	Optimize condition	Efficiency	References
H <sub>2</sub> SO <sub>4</sub> + H <sub>2</sub> O <sub>2</sub>	[H <sub>2</sub> SO <sub>4</sub> ] = 4.3 M [H <sub>2</sub> O <sub>2</sub> ] = 4.8 M Pulp density = 0.1% (w/v) Particle size = 0.5 mm	Zn = 76% Cu = 80%	[27]
HCl	[HCl] = 2 M Particle size = 1.4–3 mm <i>t</i> = 240 min <i>T</i> = 323 K	Zn = 98% Cu = 71%	[28]
<i>Bacillus megaterium</i>	pH = 10 Pulp density = 2% (w/v) Particle size = 149 μm	Cu = 13.3%	[22]
<i>A. ferrooxidans</i>	pH = 3 Fe <sup>3+</sup> = 8.4 g/L Pulp density = 20 g/L Particle size = 95 μm <i>t</i> = 80 days	Cu = 100%	[9]
Ultrasonic + H <sub>2</sub> O <sub>2</sub>	pH = 1.5 Particle size = 165 μm Ultrasonic power = 200w <i>t</i> = 90 min	Cu = 95.2%	[29]
Lemon juice + H <sub>2</sub> O <sub>2</sub>	[Lemon juice] = 74% (v/v) [H <sub>2</sub> O <sub>2</sub> ] = 12.2% (v/v) Pulp density = 1.4% (w/v) Particle size = 150–180 μm <i>t</i> = 5 h	Cu = 90% Zn = 76%	This study

**Fig. 4** SEM images of the waste particles: **a** before the bio-acid leaching process, and **b** afterwards

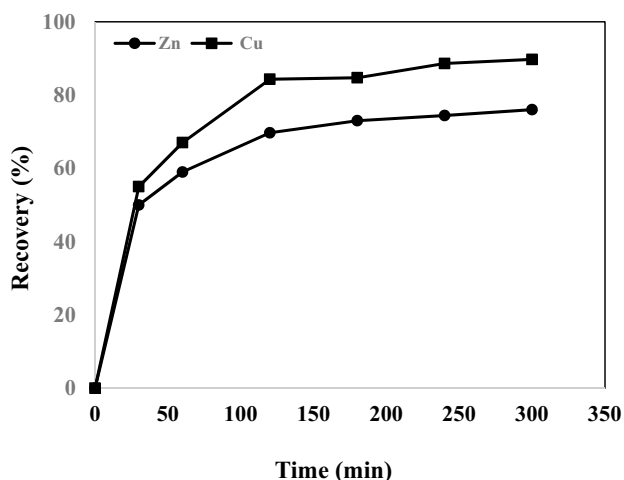
where  $X$  denotes leaching efficiency (%),  $t$  leaching time (min) and  $k$  rate constant ( $\text{min}^{-1}$ ). Figure 6 shows the plots of Eqs. 17 and 18. The  $R^2$  were 0.95 and 0.93 for Cu extraction and Zn extraction, respectively, for diffusion-controlled process. They were 0.86 and 0.85, respectively, for reaction-controlled process. Result shows diffusion is the rate-limiting step.

## Conclusion

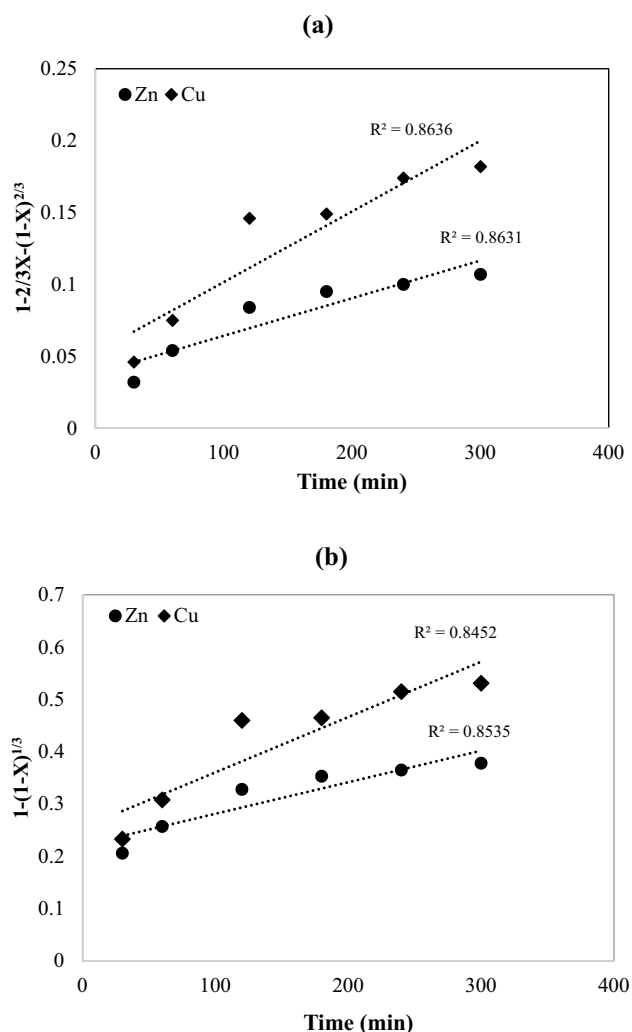
In this study, RSM was used to model Zn and Cu extraction efficiencies of PCBs using natural organic acids determined from lemon juice assisted by H<sub>2</sub>O<sub>2</sub>. Effects of lemon

juice concentration, S/L ratio, and H<sub>2</sub>O<sub>2</sub> concentration on metal extraction efficiencies were evaluated. The ANOVA results of the RSM model fitted experimental data well. The optimum leaching operation conditions were predicted to be 74% lemon juice, 12.2% H<sub>2</sub>O<sub>2</sub>, and S/L ratio of 1.41% which resulted in 89% Cu and 73% Zn recovery yields in 4 h with initial temperature of 20 °C. The effect of time on extractions of Zn and Cu metals under optimum point conditions was investigated. The kinetics results showed that after 300 min, the highest efficiencies were 90% Cu and 76% Zn and the diffusion-control model was able to model the total reaction rate. Then based on this result, the recovery of Cu and Zn from PCBs with high efficiency, low toxicity to





**Fig. 5** The effect of time on Cu and Zn recovery yields at optimal conditions



**Fig. 6** Shrinking core model: **a** with diffusion control, and **b** with chemical reaction control

environment and in short time can be done. Therefore, it can develop in pilot and industrial scale in the future.

## Declarations

**Conflict of interest** The authors declare that they have no conflict of interest.

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