Optimizing the Amino Acid Leaching Parameters of Gold from the Mobile Phone Printed Circuit Boards

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ABSTRACT: *Technological advances in electronic equipment and the variety-seeking behavior of people in using new equipment and devices have turned the issue of electronic waste (E-waste) into a major dilemma in the world. Electronic waste contains high levels of toxic metals such as lead and cadmium, base metals like aluminum and copper, precious metals such as silver, platinum, and palladium, and non-metallic parts consisting of resin, ceramic, plastic, and glass. There are different methods available to recover precious metals such as gold and platinum from electronic waste. However, the amino acid L-valine was used in this paper as a cheap and non-toxic leaching agent instead of cyanide. The grade of gold and platinum in the sample is 542 and 30 ppm, respectively, In addition, the parameters of L-valine concentration, temperature, pH, pulp solid percentage, and time were assessed by the Design of Experiments (DOE) method using the* Dx_7 *software. The experiments' results demonstrated that the parameters of temperature, time, pH, pulp density, and L-valine concentration respectively have greater importance in the gold recovery process due to the lower P and higher F values. The mean percentage of gold recovery was obtained as 81.47% when validating the results under the proposed optimal conditions (L-Valine concentration: 173 g/t, pH: 11, time: 24 hours, temperature: 80 °C, pulp solid percentage: 19%, and constant stirrer speed: 400 rpm).*

KEYWORDS*: Gold Leaching, Printed Circuit Board; L-Valine.*

INTRODUCTION

Gold is a precious metal and is widely used in various industries, including electrical and electronic industries, pharmacy, etc. due to its unique properties such as high conductivity, high corrosion resistance, flexibility, and ductility[1]. Approximately 20 to 50 million tons of electronic waste are annually generated around the world[2]. In 2016, over 630000 kilograms of electronic waste have been generated in Iran and the share of each Iranian has been about 7.6 kg; in other words, it is 1.5 kg of waste more than the global per capita[3]. The waste (electronic products waste) has been recently recognized as a major secondary source of these precious metals. The amount of gold contained in each of the electronic components of personal computers is higher than the amount of gold that can be extracted from 17 tons of gold ore. Moreover, one ton of mobile phones, which equals approximately 6,000 cell phones, contains 3.5 kg of silver, 340 g of gold, 140 g of palladium, and 130 kg of copper. The amount of copper found in the printed circuit boards of computers is estimated to be up to 400 kg per ton[1].

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The presence of highly toxic elements such as lead, mercury, arsenic, cadmium, selenium, and hexavalent chromium in electronic devices has led electronic waste to be categorized as hazardous waste, which if not organized properly will cause serious problems for the environment and human health. Therefore, e-waste recycling seems to be highly important from environmental and economic aspects[4]. The recycling process usually takes place through three main stages: Separation, shredding, and processing. The first measure to recover all types of electronic waste is to separate hazardous or valuable components. This task is done with hammers, screwdrivers, and conveyors to separate the parts into different batches for their recovery. The next step is to crush the material mechanically by crushers and mills. The crushed and milled materials are then passed through electrical and magnetic separators to separate the metal and non-metal electrical components. Ultimately, after these physical or mechanical recycling techniques are completed, hydrometallurgical and pyro metallurgical processes or a combination of processes will be employed to further refine and concentrate metals[4]. Hydrometallurgical processes are preferred over pyro metallurgical methods due to low emission of toxic and volatile gases, low energy consumption, low waste production, and easy working conditions. Some procedures should be done before hydrometallurgical extraction of metals from electronic waste, including stages of homogenization and reduction of dimensions by crushing, magnetic separation, and categorizing with sieving. Besides the cyanidation process, there are a few non-cyanide leaching processes with non-toxic solutions such as thiosulfate, thiourea, halogens, Aqua regia, and amino acids, which can form strong and stable complexes with gold, silver, and platinum to be used for dissolving precious metals[2]. The use of amino acids and hydrogen peroxide to leach gold from electronic waste may be employed as an efficient and environmentally friendly method alone or combined with other leaching agents as a gold leaching agent aimed at decreasing the consumption of leaching agent and enhancing the recovery rate of precious metals[3]. L-valine is an environmentally safe amino acid, which has no side effects on humans, animals, and plants unlike cyanide and other leaching agents. It is also economically cheaper than other leaching agents. L-valine can dissolve gold by oxidizing agents such as hydrogen peroxide and air according to Equations (1) and (2):

$$2Au + 4[NH_2CH(C_3H_7)COOH] + 2OH^- + H_2O_2 \rightarrow 2Au[NH_2CH(C_3H_7)COO]^-{}_2 + 4H_2O \quad (1)$$

$$2Au + 4[NH_2CH(C_3H_7)COOH] + 1/2O_2 + H_2O \rightarrow 2Au[NH_2CH(C_3H_7)COO]^-{}_2 + 2OH^- \quad (2)$$

Although there are different methods for leaching e-waste and the recovery of base and precious metals, researchers are still looking for cost-effective methods to enhance the recovery rate of metals meanwhile reducing environmental problems. *Deventer* (2011) studied the synergistic effect of the amino acid histidine, glycine, alanine, and valine in the thiosulfate leaching of pyrite gold concentrate and found that the synergism of amino acids reduces the consumption of thiosulfate and increases the kinetics of gold extraction. The study also demonstrated that histidine has the greatest impact on increasing the gold recovery rate and reducing thiosulfate consumption compared to other amino acids[5]. *Tripathi* (2012) evaluated leaching of gold from the waste mobile phone Printed Circuit Boards (PCBs) with Ammonium Thiosulphate. the maximum gold leaching obtained 78.8% at thiosulfate: 0.1M, copper sulfate: 40mM, pH: 10-10.5, stirring speed: 250 rpm at room temperature in 8h time duration[6]. *Birloaga* (2013) studied the synergistic effect of sulfuric acid and hydrogen peroxide on the leaching of electronic scrap and waste for the recovery of copper. In his study, a 90% recovery for copper was obtained using sulfuric acid with a concentration of 2 M in 20 mL of 30% hydrogen peroxide (for 100 mL of solution) within a period time of 2 hours at a temperature of 30 °C [7]. *Petter* (2014) evaluated the leaching process of electronic waste to recover gold by using nitric acid. A recovery rate of 100% was obtained for gold in this study using nitric acid with a volume ratio of 1/3, solid to liquid ratio of 1/20, and pre-concentration at 25 °C and 60 °C within 2 hours[8]. *Akcil* (2014) examined the synergistic effect of divalent copper, thiosulfate, and ammonia in the leaching of gold in electronic waste and obtained a recovery rate of 90% for gold under the following conditions: Thiosulfate concentration: 72.71 mM, concentration of divalent copper ions: 10 mmol, ammonia concentration: 0.266 M, pH=10, and temperature: 20 °C [2]. *Eksteen* and *Oraby* (2015) studied the process of gold leaching in sulfide minerals using amino acids at low concentrations. The experiments' results demonstrated that the rate of gold recovery 40 °C dramatically enhances with increasing temperature from to 60 °C, increasing the concentration of amino acids, hydrogen peroxide, and pH. The presence of cupric ion increased the rate of gold leaching in glycine, while the recovery rate of gold dropped in the presence of pyrite due to the consumption of hydrogen peroxide in the sulfide oxidation process[9].

Using the bacterium Chromobacterium viola, *Liu* (2016) examined the bio-leaching of gold from printed circuit boards. In this study, he used a combination of the bacterium Chromobacterium viola, iodine, and Pseudomonas aeruginosa and Pseudomonas fluorescens bacteria aimed at enhancing cyanide production, which increased the gold recovery rate to more than 70%[10]. *Tanda* (2017) studied the leaching of copper from azurite, chrysocolla, cuprite, and malachite minerals using glycine. Finally, after 24 hours under conditions of optimal pH of 11 and the glycine-to-copper ratio of 4:1, the copper leaching recovery percentage from azurite, malachite, cuprite, and chrysocolla minerals were obtained approximately as 95%, 90%, 83.8%, and 17.4%, respectively[11]. *Perea* (2018) used glycine and monosodium glutamate to examine the recovery of gold from computer components. The results indicated the high dependence of the gold recovery percentage on the parameters of the oxidant concentration and pH, which increases with the presence of cupric ions, and a 100% gold recovery rate was obtained using glycine[12]. *Cara Philipa* (2018) examined the amino acid leaching of base and precious metals from PCBs waste using glycine. The leaching of copper and other base metals was initially studied using glycine. The findings demonstrated that increasing temperature and glycine concentration in the presence of air enhances the dissolution rate of copper. Then, the precious metal leaching tests were done by utilizing the cake left over from the base metal leaching experiments. Two leaching stages were made with a duration of 41 to 52 hours to obtain a 78% copper recovery rate. Finally, a recovery rate of about 38% was obtained by gold leaching under the conditions of 0.04 M sodium cyanide concentration and 0.13 M glycine concentration at 25 °C and 96 hours with an air oxidizer [13]. Using the experiments design method, *Aghababaei* (2017) investigated effective parameters in copper leaching from PCBs waste by sulfuric acid and hydrogen peroxide such as sulfuric acid concentration, acid-to-oxidizer ratio, temperature, time, and solid percentage. Ultimately, a recovery rate of 96% was achieved for copper under the conditions of 4 M sulfuric acid concentration, acid to oxidizer ratio of 4, the temperature of 50 °C, time of 4 hours and 35 minutes, and a solid percentage of 5%, and the stirring speed of 300 rpm[14].

Raeisi (2019) studied the extraction of gold from the electronic boards of used computers by the solvent extraction method. The leaching of such waste was initially done using the Aqua regia solution. Then, the trioctylamine extraction agent was used in the solvent extraction process to extract the gold in the solution resulting from the leaching process. He used the design of experiments approach by Response-Surface Methodology (RSM) to optimize the parameters influencing the gold leaching process, including the extractor concentration, aqueous phase to organic phase ratio, hydrochloric acid concentration, gold concentration in the stock solution, and time. Finally, 99.6% of gold and 23.4% of copper contained in the prototype were transferred to the organic phase[15]. *Kiani* (2019) utilized amino acids together with cyanide and alone in the cyanation process of the Poyazarkan Agh Dareh gold processing plant. According to the results, the recovery rates for gold, silver, and mercury were obtained as 90%, 55%, and 19%, respectively, under the conditions of 300 g/te concentration, ambient temperature, pH of 10.5, time of 24 hours, pulp density of 40%, the particle size of 53 microns with a constant stirring speed in a cyanide leaching process, but under the conditions of 300 g/te concentration, the temperature of 75 °C, pH of 11.5, time of 24 hours, pulp density of 30%, and particle size of 53 microns, the recovery rates for gold, silver, and mercury were respectively achieved for the amino acid creatine monohydrate as 91%, 47.18%, and 0%, for the amino acid L-valine as 90.5%, 16.56%, and 0%, for the amino acid L-histidine as 90.5%, 15.92%, and 0%, respectively[16]. *Tuncuk* (2019) examined the selective leaching of precious metals from memory rams waste in two stages. In the first stage leaching with concentrations of 2% iodine and 3% hydrogen peroxide, a pulp density of 5%, and a time of 2 h, 81.81% and 99.02% of gold and silver were recovered, respectively.

About 79.3% silver was recovered in the second stage with sulfuric acid with a concentration of 2 M, 1.5 mg concentration of ammonium thiosulfate, pulp solid percentage of 5%, and time of 5 hours. Ultimately, 73.98%

Fig. 1: ***Primary and secondary crushing of the representative sample and dimensions of the crushed sample.***

of copper, 99.98% of gold, and 90.96% of silver were recovered after two leaching stages[17]. *Gang Zhou* (2020) studied the recovery of gold from mobile PCBs waste using the bioleaching method and by cyanide-producing bacteria of Pseudomonas putida and Bacillus megaterium. Finally, a gold recovery rate of 87.46% was obtained at a pH of 10, the pulp solid percentage of 5 g/l, and the time of 34 hours[18]. *Wang* (2021) studied recycling gold from printed circuit boards gold-plated layer of waste mobile phones in "mild aqua regia" system. In this study, gold was recycled without pretreatments or enrichment processes from the gold-plated layer on the PCBs in WMPs, by using a DMF-$CuCl_2$-$CaCl_2$ reaction system called "mild aqua regia". the developed system has good selectivity of gold over base metals, and under the optimized conditions, leaching and precipitation rates of gold was obtained over 99%[19]. *Salinas-Rodríguez* (2022) studied leaching of Copper contained in waste printed circuit boards, using the Thiosulfate Oxygen System. According to the results, it was found that in the studied system of $S_2O_3{}^{2-}$ and O_2, the leaching of copper with values of Ea = 25.78 kJ/mol and n = 0.22 (for the leaching reagent), reaction was controlled by the oxygen transport to the solid–liquid interface and also by the chemical reaction in the surface of particles, was obtained up to 99.82% copper in solution[20].

The process of recovering gold from mobile phone PCBs is an innovative approach to obtain the precious metals Literature reveals that only limited studies have been carried out to recover the metals from waste mobile phone PCBs. So far, the amino acid leaching process (especially by glycine) alone or combined with cyanide and other leaching agents has been studied mainly for the recovery of metals from ores or gold and silver sheets. Hence, the use of amino acids, especially L-valine in gold leaching from electronic equipment waste in the world has been less studied than other leaching methods as a gold leaching approach and no research has been conducted in Iran in this area so far. Accordingly, there are still unknowns regarding the use of these materials in the precious metals leaching process. Therefore, in this study, the amino acid L-valine was used as an environmentally friendly method and a new leaching agent and also a proper alternative for cyanide to recover gold from the mobile phone printed circuit boards, and the influential parameters were evaluated by the design experiments method using the Dx_7 software In this study, the experimental data are analyzed statistically using Design Expert software (Demo version 7.0.0, from Stat-Ease Inc., Minneapolis, MN, USA).

EXPERIMENTAL SECTION

Preparation and crushing of the representative sample

In the first step, a mixture of old and new printed circuit boards was prepared from the Abdol Abad neighborhood of Tehran and the Motahhari neighborhood of Khorramabad. Then, the preparation operation was done manually and the ceramic, plastic, and metal parts were removed from the circuit boards. Afterward, the crushing operation took place in two stages using the hammer mill and pin mill related to the Danesh Faravaran Company located in Parand Industrial Town of Tehran, and the dimensions of the sample were reduced to less than one millimeter.

Sieve analysis

The dimensional distribution of the representative sample was done by dry sieve analysis using standard sieving series American Society for Testing and Materials (ASTM), respectively as 16-130-45-60-60-80-100-140 meshes. Then, according to the plotted sieve analysis diagram (Fig. 2), the d_{80} of the representative sample was calculated as 684 microns.

Table 1: The atomic absorption spectroscopy analysis

Element Name	Au	Pt	Pd
Fineness (ppm)	542	30	26

Table 2: The inductively coupled plasma (ICP) analysis

Element Name	Ag	Al	As	Ba	Be	Bi	Ca	Cd	Ce	Co	Cr
Weight Percent	0.17	1.02	0.004	0.725	0.003	0.008	1.415	0.001	0.003	0.008	0.325
Element Name	Cu	Fe	K	La	Li	Mg	Mn	Mo	Na	Hg	
Weight Percent	30.675	3.505	0.021	<0.001	0.001	0.191	0.066	0.005	0.039	<0.001	

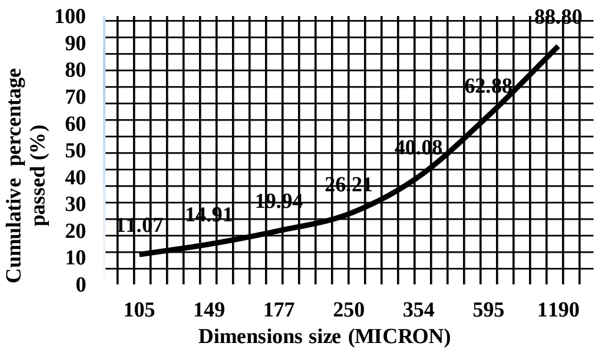


Fig. 2. The sieve analysis diagram of the representative sample

Atomic Absorption Spectroscopy (AAS) and Inductively Coupled Plasma (ICP) spectroscopy

The fineness of the base and precious metals in the PCBs of mobile phones was assessed and determined with two methods of Atomic Absorption Spectroscopy (AAS) and Inductively Coupled Plasma (ICP) Spectroscopy by ZarAzma Mineral Studies Company. The analysis results of precious metals such as gold and platinum by the AAS method and the analysis results of base metals are given in Tables 1 and 2, respectively (100 grams of homogeneous sample was send to the laboratory for atomic absorption analysis and ICP analysis). As seen in Tables 1 and 2, the waste used contains significant amounts of precious metals such as gold, platinum, palladium, and base metals.

Materials and equipment

Chemicals from Merck Co., Germany, such as sulfuric acid (with a purity of 98% as a copper leaching agent), amino acid L-valine (with a purity of 99% as a gold leaching agent), hydrogen peroxide (30% as the oxidizer), and sodium hydroxide (with a purity of 99% as a pH regulator) were used. A model magnetic stirrer (HPMA 700) was utilized to control the temperature and stir the pulp; also, a pH meter of SITEC company available in Lorestan University's Mineral Processing laboratory was employed to measure pH. Finally, an American-made Agilent Atomic Absorption Spectroscopy, model (AAFS 240) available in the central laboratory of Lorestan University was used for analyzing the gold and copper content of the dissolved and waste samples obtained from the leaching process.

Leaching tests

The leaching experiments were conducted on 10-g representative samples. A pretreatment stage was performed with sulfuric acid as a leaching agent and hydrogen peroxide by a magnetic stirrer equipped with a temperature controller prior to conducting leaching tests with the amino acid L-valine considering the high percentage of copper in the mobile PCBs (30.675%). The acid washing operation using sulfuric acid was done under optimal conditions (4 M sulfuric acid concentration, acid to oxidant ratio of 4, a temperature of 50 °C, a time of 5 h, and a solid percentage of 5%). Ultimately, 68.4% of the copper content in the PCBs of mobile phones was removed in form of a solution. Then, 24 amino acid leaching tests were designed and performed using L-valine on the cake obtained from acid leaching by experiments design method and using the Dx_7 software.

Designing the leaching test

After acid leaching of 10-g samples by sulfuric acid and the removal of 68.4% of the copper content in the PBCs as a solution. the leaching tests were performed using L-valine amino acid. After identifying the effective factors and the range of their variation, five important factors, including L-valine concentration, temperature, pH, time, and the pulp density were selected for modeling and optimizing the factors influencing the L-Valine

Table 3: The selected levels of factors in terms of actual values and code based on the central composite design

Factors	Sign	Low Factorial Level (-1)	High Factorial Level (+1)	-α	+α
Concentration of L-valine (g/t)	A	150	250	100	300
pH	B	10	12	9	13
Time (hour)	C	20	30	15	35
Temperature (degrees Celsius)	D	60	80	50	90
Pulp density (%)	E	15	25	10	30

leaching process. Then, the experiments were designed using Dx_7 (Design Expert) software by the surface-response (central composite design) method, and experiments were performed on the representative samples (mobile PBCs) under different conditions. The effective factors associated with their experimental values are given in Table (3).

Also, the design matrix of the experiments performed by combining different levels of the examined parameters (24 experiments) and the calculated values of the response variable of each test (gold recovery) are provided in Table (4). As seen, the mean gold recovery rate was obtained as 82.8% under the optimal conditions of L-valine concentration of 150 g/t, a temperature of 80 °C, the time of 30 hours, oxidizer amount of 3 mL, pulp density of 15%, and the pH of 12.

Analysis of variance

The analysis of the variance of laboratory data was made by a series of mathematical operations aimed at identifying the effective factors and their order of importance. The results of the analysis of variance are given in Table 5.

The results provided in Table 5 can be used to validate the proposed model. As seen, the correlation coefficients of R^2 and Adjusted R^2 were obtained as 0.9815 and 0.9392, respectively, suggesting the efficiency of the model. Adequate precision is another criterion to assess the model, which is the signal-to-noise ratio criterion and its optimal value needs to be higher than 4. In this case, an adequate precision of 15.773 was achieved, which indicates a sufficient signal, and thereby, the suitability of the model to conduct the design space. The results in Table 5 suggest that the parameters of temperature, time, and pH respectively matter more in the gold recovery process than pulp density and L-valine concentration due to lower P-value and higher F-value. Moreover, the mathematical model provided by the software was proposed according to Equation (2) to predict the gold recovery rate.

$$RAu=77.28-3.81\times A+3.8\times B+4.48\times C+6.03\times E+3.05\times A\times B+5.02\times A\times E-3.4\times B\times D+3.82\times B\times E-5.47\times C\times D-1.98\times D\times E-1.98\times A^2-1.6\times B^2-1.03\times C^2-2.28\times D^2 \quad (2)$$

Analysis of modeling errors

We had to examine the residuals (differences between the values obtained by the model and the values measured in the experiments) to assess the accuracy of the fitted regression equation in predicting the values of the response variable (gold recovery) for different levels of factors. Fig. 3 shows the comparison of actual recovery values with the predicted recovery values; the closer these values are to the straight line, the closer the answer predicted by the software will be to the actual answer.

RESULTS AND DISCUSSION

Evaluating the effect of factors on the gold leaching recovery

In this section, the effect of each parameter was examined individually and in interaction with important parameters.

The effect of L-valine concentration

As illustrated in Fig. 4, the gold recovery percentage drops from about 78% to 67% by increasing the concentration of L-valine from 150 to 250 g/t. When the concentration of L-valine in the solution increases, these compounds react with themselves to form a peptide (amide) bond. In fact, peptide bond formation may act as one of the core factors in reducing the percentage of gold leaching recovery by increasing the amount of L-valine[12]. In addition, at alkaline pH, the increased concentration of L-valine will enhance the rate of amino acid reaction with copper oxide and other base metals, and thereby, the amount of L-valine effective in dissolving and recovering gold would decrease[13].

Table 4: The central composite design matrix to perform tests and the values of the answers (gold recovery)

Test Number	A	B	C	D	E	Gold Recovery (Percent)
1	200	11	25	70	10	76.35
2	300	11	25	70	20	59.96
3	250	10	30	80	15	72.49
4	100	11	25	70	20	76.75
5	200	11	25	90	20	79.6
6	150	12	30	80	15	82.8
7	150	12	30	60	25	77.79
8	200	11	25	50	20	54.75
9	150	12	20	80	25	70.82
10	200	11	25	70	20	77.3
11	200	11	15	70	20	63.58
12	150	10	20	60	15	64.4
13	150	10	30	80	25	65.5
14	250	10	20	80	25	64.5
15	250	12	20	80	15	72.58
16	200	9	25	70	20	62.54
17	200	11	25	70	30	63.93
18	250	12	20	60	25	67.10
19	200	11	35	70	20	80.76
20	200	11	25	70	20	77.8
21	200	11	25	70	20	78.7
22	250	12	30	60	15	74.09
23	200	13	25	70	20	77.27
24	250	10	30	60	25	59

Table 5: The Results of the analysis of variance using the response level method for gold recovery in the L-valine leaching

Source of changes	Sum of squares	Degree of freedom	Mean of squares	F-value	P-value	Description
Model	1425.48	16	89.09	23.21	0.0002	Significant
A- Concentration (L-valine)	184.55	1	184.55	48.07	0.0002	
B-pH	215.64	1	215.64	56.17	0.0001	
C- Time	220.93	1	220.93	57.55	0.0001	
D-Temperature	399.50	1	399.50	104.06	<0.0001	
E-Solid Percent	197.86	1	197.86	51.54	0.0002	
AB	46.93	1	46.93	12.22	0.0100	
AE	127.64	1	127.64	33.25	0.0007	
BD	82.86	1	82.86	21.58	0.0024	
BE	83.18	1	83.18	21.67	0.0023	
CD	122.57	1	122.57	31.93	0.0008	
DE	27.98	1	27.98	7.29	0.0307	
A^2	93.33	1	93.33	24.31	0.0017	
B^2	60.42	1	60.42	15.74	0.0054	
C^2	25.15	1	25.15	6.55	0.0376	
D^2	123.15	1	123.15	32.08	0.0008	
E^2	56.05		56.05	14.60	0.0065	
Residual	26.87	7	3.84			
Lack of fit	25.87	5	5.17	10.28	0.0910	Insignificant
Pure error	1.01	2	50.50			
Standard Deviation	Mean	C.V (%)	R^2	Adjusted R^2	Predicted R^2	Adequate Precision
1.96	70.85	2.77	0.9815	0.9392	0.6997	15.773

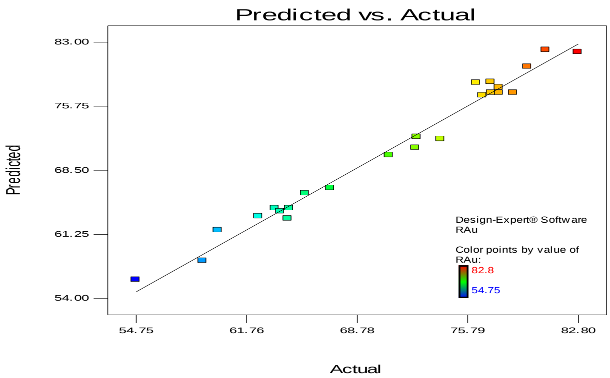


***Fig. 3:** The diagram of gold leaching recovery laboratory values versus values predicted by the model (R-square=0.9815)*

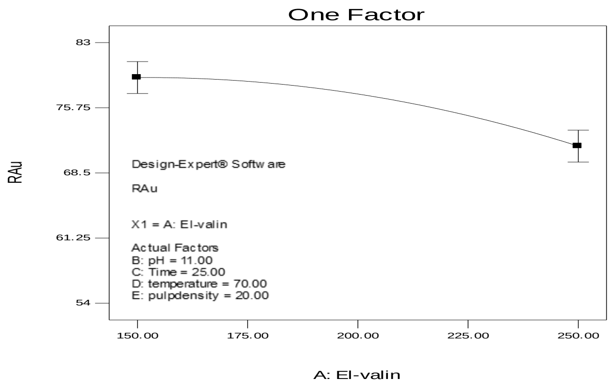


***Fig. 4:** Effect of L-valine concentration on leaching of gold (pH: 11, temperature.: 70°C, pulp density: 20, time: 25h)*

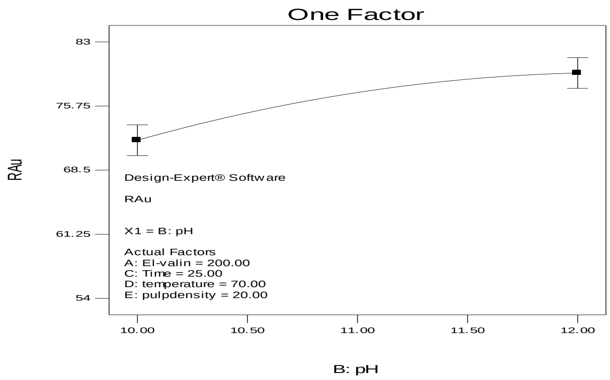


***Fig. 5:** Effect of pH on leaching of gold (L-valine concentration: 200 g/t, Time: 25h, pulp density: 20, temperature: 70°C)*

The effect of pH

As seen in Fig. 5, the gold recovery percentage increases from about 70% to 81% as a quadratic function by increasing the pH from 10 to 12, and the slope of the curve gradually decreases at higher pHs. Sodium hydroxide was used in leaching experiments to adjust the pH. As realized in Equation (3), hydrogen peroxide decomposes with alkali at high pHs.

$$H_2O_2 + OH^- \leftrightarrow H_2O + HOO^- \quad (3)$$

The decomposition of hydrogen peroxide to produce oxygen and hydroxide ions increases at higher pHs (10 to 11), as shown in Equation (4).

$$HOO^- + H_2O_2 \rightarrow H_2O + OH^- + O_2 \quad (4)$$

Further enhancement of pH (11 to 12) and leaching temperature can cause the production of hydroxide ions from hydrogen peroxide according to Eq. (5).

$$H_2O_2 \rightarrow 2\ OH^- \quad (5)$$

The generated hydroxide ions can enhance the leaching rate and increase the gold leaching recovery rate[21]. On the other hand, once the pH increases (11 to 11.5), the hydrogen is taken from the amino acid group, and a negative charge is formed on the amino acid so, causing the amino acid reacts more easily with the metal, and thereby, the leaching recovery will be increased. Then, by increasing the pH, the hydrogen of the amino group is taken and this compound will turn into a strong nucleophilic compound, which can react with any electrophilic compound in the solution. As a result, the amount of effective L-valine decreases, and ultimately, the recovery rate of gold leaching would decrease[16].

The effect of time

As illustrated in Fig. 6, the gold leaching time has varied between 20 and 30 hours with the percentage of gold recovery changing between 70% and 82%. Based on the experiments' results, one can predict that the percentage of gold leaching recovery will be on the rise with increasing time.

The effect of temperature

According to the results seen in Fig. 7, increasing the temperature between 60 °C and 80 °C would enhance the percentage of gold recovery from about 68% to 82% as a quadratic function. Also, the slope of increasing gold recovery rate over time is greater compared to other examined parameters. In other words, increasing the temperature would enhance the rate of gold leaching with the amino acid L-valine, and ultimately, increases the gold recovery rate.

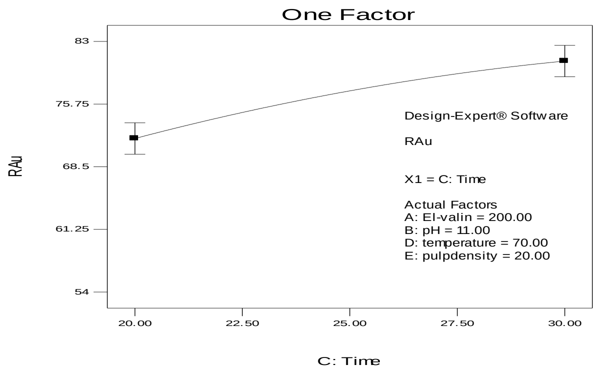


Fig. 6: Effect of time on leaching of gold (L-valine concentration: 200 g/t, pH: 11, pulp density: 20, temperature: 70°C)

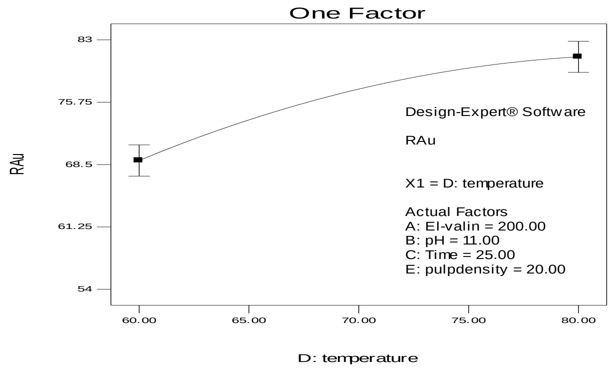


Fig. 7: Effect of temperature on leaching of gold (L-valine concentration: 200 g/t, pH: 11, pulp density: 20, Time: 25h)

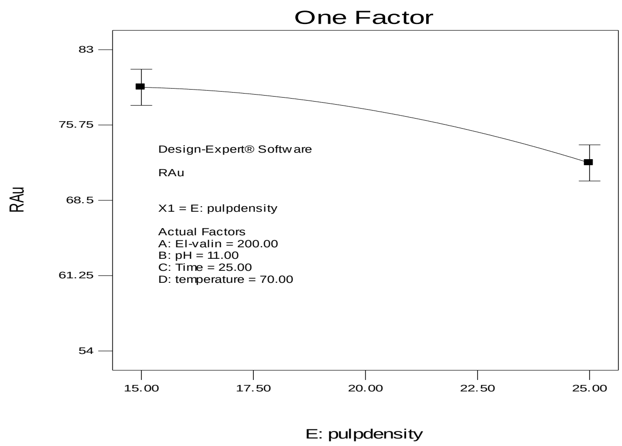


Fig. 8: Effect of pulp density on leaching of gold (L-valine concentration: 200 g/t, pH: 11, Time:25h, temperature: 70°C)

The effect of pulp density

As can be seen in Fig. 8, the gold recovery rate is reduced from about 80% to 70% by enhancing the pulp density from 15% to 25%. Moreover, the recovery of gold at the 25%, and 15% solid pulp is at the lowest and highest levels, as a result respectively.

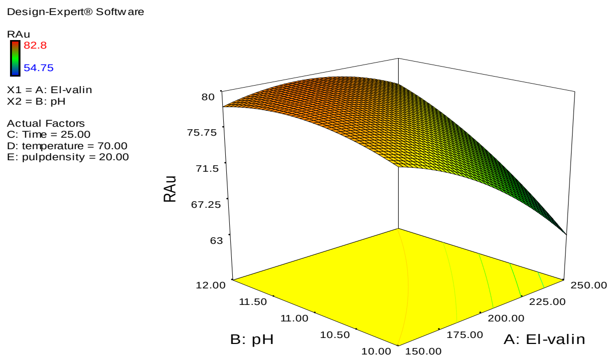


Fig. 9: The 3D diagram of the effect of interaction between L-valine concentration and pH on the gold recovery

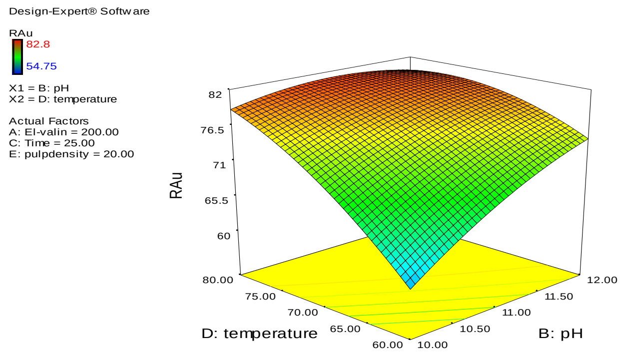


Fig. 10: The 3D diagram of the effect of pH and temperature interaction on the gold recovery

This may occur due to the reduction of oxygen transfer to the gold surface of an increase in the pulp density, which leads to a reduction in the gold leaching recovery rate[22].

Examining the effect of parameters interaction

Examining the effect of interaction between L-valine concentration and pH

As seen in Fig. 9, when the pulp pH is at its lowest value, i.e., 10, the percentage of gold recovery decreases by 10-15% by enhancing the L-valine concentration. However, when the pulp pH is at its highest value, i.e., 12, increasing the L-valine concentration does not cause much change in the percentage of gold recovery. This confirms the higher efficiency of the amino acid L-valine at pH above 10 for gold recovery.

Evaluating the effect of pH and temperature interaction

According to Fig. 10, when the temperature is at its lowest value, i.e., 60 °C, increasing the pH will enhance the gold recovery rate more than the time at which the temperature is at its maximum value, i.e., 80 °C.

Table 6: The validation of the optimal conditions proposed by the software

Experiment Number	L-valine concentration (g/t)	pH	Time (hours)	Temperature(°C)	Pulp density	Predicted recovery (%)	Actual recovery (%)	Relative difference
1	173	11	24	80	19	84.06	80.96	3.6
2	173	11	24	80	19	84.06	83.24	0.97
3	173	11	24	80	19	84.06	82.13	2.29
Mean							81.47	2.59

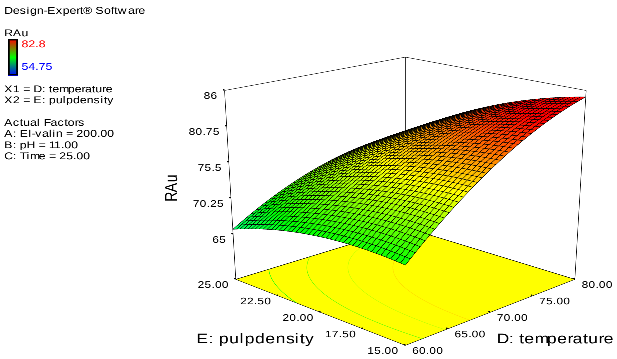


Fig. 11: The 3D diagram of the effect of interaction between temperature and pulp density on the gold recovery

Evaluating the effect of interaction between temperature and pulp density

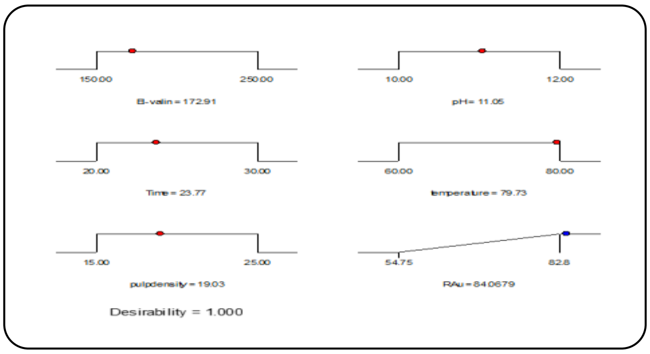
As can be seen in Fig. 11, when the pulp density is at its lowest value, i.e., 15, enhancing the temperature will increase the percentage of gold recovery by 10-15%. Also, when the pulp density is at its maximum value of 25, enhancing the temperature will increase the percentage of gold recovery by 5-10%. In other words, at a solid percentage of 15%, increasing the temperature has a greater impact on the enhancement of the gold recovery rate.

Optimizing the parameters influencing gold leaching by L-valine amino acid

The operating conditions of each parameter were optimized by Dx_7 software with the terms of the maximum gold recovery percentage and keeping other values of the parameters in the predetermined range. The results of the optimization made by the software are given in Fig. 12. Three experiments were conducted with the optimal conditions suggested by the software to validate the test design results. Table 6 shows the conditions of the experiments associated with the comparison of the predicted and actual results of the gold recovery process. According to the results, the best gold recovery rate was obtained in practice as 83.24% with a concentration of 173 g/t, a pH of 11, a time of 24 hours, a temperature of 80 °C, a pulp density of 19%, and the constant stirrer speed of 400 rpm.

CONCLUSIONS

Electronic waste is seen as a treasure trove of base and precious metals. The presence of a high percentage of copper in the electronic waste of the examined mobile phone (30.675%) enhances the consumption of the leaching agent, and thereby, increases the cost of the leaching process. Thus, 68.4% of the copper content in the PCBS was initially removed in the form of a solution by doing an acid leaching stage with sulfuric acid. This reduced the consumption of the amino acid and decreased the relevant costs. Also, the gold alloyed with copper metal was placed in better conditions for leaching with the amino acid, and thereby, the gold recovery rate increased by about 25.11% compared to the leaching process with L-valine without the acid washing stage. Moreover, the results of the experiments design method demonstrated that the parameters of temperature, time, and pH respectively matter more than the parameters of pulp density and L-valine concentration in recovering gold from mobile PBCs wastes. The amino acid-assisted leaching experiments were performed by the experiments design method using the Dx_7 software. Finally, the mean gold recovery rate was obtained as 83.24% under the following optimal conditions: L-valine concentration: 173 g/t, pulp density: 19%, and pH: 11. As a comparison between the present study and the studied conducted in 2021 by Wang on mobile phone circuit boards for gold recovery using Aqua regia, the following results can be obtained. In the present study, after performing a pretreatments step and removing a high percentage of copper and other base metals, a high recovery for gold was obtained by amino acid L-valine and also is cheaper and more compatible with the environment than other methods. While, Aqua regia can be used cyclically without pretreatments or enrichment process,



ig. 12: The picture graph of the optimal conditions of the gold leaching parameters

and the reaction system could be cyclic utilized. In Aqua regia system, gold can be recovered through leaching, precipitation and filtration. It is an efficient method to recover precious metals from scrap electrical and electronic equipment, but this method can bring a lot of cost and environmental and human risks [19].

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