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Recovery of Copper and Gold from Waste Printed Circuit Boards Using Monosodium Glutamate Supplemented with Hydrogen Peroxide

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Abstract: This study aimed to recover copper and gold from a waste printed circuit board (WPCB) using monosodium glutamate (MSG) supplemented with a clean oxidizer(s) under 30 °C, 150 rpm, and an initial pH of 7.00. The solder mask of WPCBs needed to be removed before leaching. At the first leaching, 93% of copper was selectively leached from solder–mask–free WPCBs within 3 h using 0.75 M MSG with 0.50% (w/v) H_2O_2. Then, gold (86%) was extracted from the first residue for 3 h by 1 M MSG with 0.25% (w/v) H_2O_2. In the downstream process, 98–100% copper could be directly recovered within 5–6 h from the first leachate by electrowinning (EW). However, gold in the second leachate required separation and purification processes before EW. At 4 h, 98% gold was adsorbed onto the activated carbon. Most impurities were removed from gold-loaded activated carbon by pre-elution using 0.05 M EDTA for 0.5 h without gold release. Afterward, 99% gold was eluted by diluted aqua-regia within an hour. Finally, the gold could be harvested perfectly using EW for 2 h. This complete copper and gold recycling process delivers an innovative concept for hydrometallurgical study and has the potential to develop into a practical industry.

Keywords: leaching; adsorption; elution; electrowinning

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1. Introduction

Electronic wastes (e-wastes) are various electrical devices and components that are undesired and discarded due to expiration and obsolescence. E-waste generation has thus far increased enormously. The amount of e-waste is three times that of other types of municipal waste [1]. Worldwide, Saha et al. (2021) indicated that up to 53.6 million metric tons (MMT) of e-waste was produced internationally in 2019 [2], and this value increased to 57.4 MMT in 2021 [3], of which approximately 24.9 MMT was generated mostly from Asia. Expectedly, global e-waste is predicted to reach 74.7 MMT in 2030 because of the rapid growth of the economy and technology as well as the lifestyle trends of the new generation. E-wastes are usually considered secondary ore and have received a lot of attention from metallurgists because of their high metal content and great metal diversity [4], including base metals, such as copper and iron, precious metals, such as gold and platinum, and hazardous metals, such as lead and chromium [5]. However, only 20% of global e-waste was formally recycled, and the remaining 80% was frequently incinerated or disposed of in landfills. To obtain the economic metals and reduce hazardous substances before disposal, e-waste recycling is necessary.

The waste printed circuit board (WPCB) is one of the major e-wastes. They are composed of a copper-clad laminate, which is coated by the solder mask layer to prevent oxidation. Following Zhou and Qiu (2010), the proportion of WPCBs in e-wastes was estimated to be approximately 3%. WPCBs comprise two main parts: (1) a non-metal part (>70%), such as plastic, resin, and glass fibers, and (2) a metal part, mainly copper [6]. Gold is one of the highly valuable metals that is present in WPCB, as mentioned above. However, the gold content in WPCB is variable. It is strongly dependent on several factors, such as the type, source, model, and year of manufacturing [7]. Currently, the extraction of various metals from WPCBs has been extensively studied from the laboratory to industrial scales.

The hydrometallurgical process has been widely utilized in the extraction and recovery of metals from WPCBs through the use of chemicals or microbial metabolites that convert insoluble metal into a soluble form. Nevertheless, most of the substances that are presently used are harmful to the environment and human health, such as cyanide and strong acids. After leaching, the recovery process was implemented to harvest soluble metals for the next several applications.

Environmentally friendly, safe, and cost and time-effective leaching using amino acids has been recently used as a replacement for conventional extraction approaches. A variety of research has demonstrated clearly that the amino acid plays a key role as an optional chelator to complexes with metals, resulting in the dissolution of metals with high efficiency. Table 1 shows the studies that have been involved in the extraction of metals. The authors classified the amino acids used in the leaching process into three main groups according to the amino acid sources: (1) pure amino acids [8–13], (2) microbial amino acids [14–16], and (3) monosodium glutamate (MSG) [17,18].

Table 1. Some reported studies involving the extraction of metals using amino acid-based solutions.

Lixiviant	Oxidizer	Additive	Sample	Leaching (%)	Time	Initial pH	Temp. (°C)	Pulp Density (%)	Reference
1. Pure amino acids									
0.5 M Glutamate	0.03 $KMnO_4$	-	Pre-treated computer pin	11 (Au)	24	9.4	25	-	[8]
1.5 kg t^{-1} glycine	3 kg t^{-1} $KMnO_4$	-	Gold oxide ore	85 (Au)	48 h	10.5	RT [a]	30	[9]
1.25 M glycine	-	1 L min^{-1} O_2	Gold ore	90 (Au)	24 h	12	60	10	[10]
0.5 M glycine	2% H_2O_2	500 ppm NaCN	Polymetal gold ore	90 (Au) 27 (Cu)	72 h	11	RT [a]	30	[11]
		200 ppm NaCN		80 (Au) 10 (Cu)					
750 µM glycine 750 µM aspartic acid 750 µM glutamic acid 750 µM lysine 750 µM tryptophan	-	-	Silicate ore	34 (Au) 41 (Au) 40 (Au) 36 (Au) 26 (Au)	15 days	7	30	4	[12]
20 g/L glycine	10 g L^{-1} $KMnO_4$	20 g L^{-1} carbon	Flotation tailings	77 (Au) 65 (Cu)	48 h	10.5	RT [a]	30	[13]
2. Microbial amino acids									
Bacillus sp. and *Pseudomonas* sp.	-	-	Gold-bearing oxide ore	68 (Au) 54 (Ag)	20 days		Heap leaching		[14]
Heterotrophic bacteria	-	Thiosulfate and copper ions	Pre-treated polymetallic sulfide ore	79 (Au) 70 (Cu) 59 (Ag)	30 days		Heap leaching (pH: 9.5–10.0)		[15]
Growth supernatant of *Macrococcus caseolyticus* and *Acinetobacter calcoaceticus*	-	-	Silicate ore	40 (Au)	15 days	7	30	4	[16]
3. Monosodium glutamate (MSG)									
1 M MSG	-	-	Electric arc furnace dust	86 (Cu)	4 h	9	30 55 80 (Minor impact)	5	[17]
				99 (Zn)	2 h				
0.5 M MSG	0.03 M H_2O_2	-	Pins of computer	92 (Cu)	2 h	9.44	RT [a]	0.039	[18]

[a] Room temperature.

Although high metal extractability was achieved using pure amino acids, the main disadvantage of these amino acids is their extremely expensive price. Likewise, the cultivation of microorganisms that produce amino acids is cost-intensive, particularly considering the technical equipment and medium. In addition, aseptic processing and long-time cultivation are also major obstacles in the use of microbial amino acids. A few years previously, researchers started to utilize glutamate from MSG in the extraction of metals, especially copper and zinc, as shown in Table 1.

Monosodium glutamate is an easily available, economical, environmentally friendly, and biodegradable amino acid product. Moreover, over 90% can be reused by acidifying the leachate to a pH of three [17]. All previous publications attained the dissolution of metals in an alkaline pH, whereas studies on metal leaching in a neutral pH were rarely available. Hence, the extraction of copper and gold from WPCBs under pH 7.00 was studied in this research.

Powerful oxidizers (such as H_2O_2, $KMnO_4$, $KClO_3$, and $NaClO_3$) are extremely important and are widely used to enhance leaching. According to Tauetsile et al. (2018), gold glycinate can be formed under a neutral to alkaline pH with a favorable temperature (23–60 °C) in the presence of an oxidizer [19]. Li et al. (2022) also reported the requirement of an oxidizer for the conversion of zero-valent metals to a higher valence for binding with glycine [20]. Nevertheless, most of them are harmful to the environment and lead to risks in terms of health and security. Furthermore, the products of some oxidizers, such as $KClO_3$, $NaClO_3$, and $KMnO_4$ have significant effects on the downstream process, especially inorganic salts and acid residues [21]. This is one of the challenges that must be regarded in future larger-scale developments and applications. On the other hand, H_2O_2 and O_3 are cleaner, safer, and more suitable oxidizers for metal leaching due to their final products, which are water (H_2O), oxygen (O_2), and hydroxide ions (OH^-) [22]. Therefore, in this study, H_2O_2 and O_3 were supplemented separately or in combination with an MSG solution.

Due to the high effectiveness and environmental friendliness of H_2O_2, it is widely used as a chemical substance in a wide range of applications, such as wastewater treatment, dental bleaching, medical disinfection, and mining. It is greatly unstable and easily decomposes (sharply decomposes when the pH is over five). There are many factors involved in the acceleration of H_2O_2 decomposition, i.e., UV light, pH, temperature, and transition metals [23]. H_2O_2 is a weakly acidic chemical and can act as an oxidizing agent in both acidic and basic media. The metal leaching in the presence of H_2O_2 is shown in Equation (1) [22]. It can be explained that the electron(s) from metals were electrochemically received by H_2O_2 to convert the zero-valent metals to soluble metals and produced the hydroxyl radical ($^{\bullet}OH$) and OH^- as byproducts. Furthermore, $^{\bullet}OH$ also serves as a strong oxidizer for further metal oxidation [10,24]. The reaction between metals and hydroxyl radicals was reported by Jadhav et al. (2015) [22] and is presented in Equation (2):

$$M\ (s) + H_2O_2\ (aq)\ \rightarrow\ M^{2+}(aq) + {}^{\bullet}OH\ (aq) + OH^-\ (aq) \tag{1}$$

$$M\ (s) + {}^{\bullet}OH\ (aq)\ \rightarrow\ M^{2+}(aq) + OH^-\ (aq) \tag{2}$$

where M = metal(s).

The metals mounted on WPCBs are coated by a polymeric transparent thin film, including epoxy coating or organic coating (solder mask). They play a huge role in the protection of boards from chemical corrosion [25]. The presence of a solder mask is responsible for low metal dissolution due to the low penetration of lixiviant [22]. To eliminate the solder mask layer, several techniques have so far been developed and studied. Conventional solvent stripping by NaOH is mostly used to remove the solder mask of WPCBs in much research associated with metal extraction because of the high removal efficiency [26–28]. Therefore, the extractions of copper and gold from WPCBs and NaOH-pretreated WPCBs (solder-mask-free WPCBs) were compared in this current study.

To achieve the high leaching potential of precious metals, the base metals are necessarily dissolved, particularly copper (the highest-concentration metal). Li et al. (2022) and Oraby et al. (2020) reported using the two-stage leaching and glycine-based solution. The base metals were selectively leached at the first stage, whereas the precious metals were then extracted from the solid residue of the first stage (the second leaching) [9,13].

This behavior is largely due to the standard reduction potential (E^0) of base metals, which are commonly higher than that of precious metals, leading to easier oxidation. In this research, two-stage leaching was consequently performed to recover copper and gold from solder-mask-free WPCBs. The extractions of copper and gold were used as representatives of the extractability of the base metal and precious metals in the first and second leaching using MSG-based lixiviant, respectively.

Subsequent to the leaching process, a downstream process is required for the separation, purification, and recovery of metals. One of the popular methods used for the separation of copper is solvent extraction which is the use of commercially available and selective reagents such as LIX and ACORGA [29]. After the separation of copper from other impurities, copper recovery is required to obtain a zero-valent metallic copper product. There are many approaches for recovering copper, such as precipitation and electrowinning (EW). However, relatively few studies have explored copper recovery from amino acid leachate. In this study, EW was chosen to directly recover copper from the first leachate without the separation process, as it is cost-effective, easier, more sustainable, and eco-friendlier. In addition, EW is a widely used approach in several mines and industries [30].

From the 1980s until now, gold separation through the adsorption of activated carbon has been most widely utilized in numerous gold mines [31]. Activated carbon has a high potential to adsorb gold glycinates, both in the presence or absence of cyanide [32]. The gold adsorption using activated carbon can be divided into three dominant processes, including (1) the carbon-in-pulp (CIP), which is the leaching and adsorption of gold sequentially, and (2) the carbon-in-leach (CIL), which is leaching and adsorption of gold simultaneously, and (3) the carbon-in-columns (CIC), in which a pregnant leach solution (PLS) is run through the activated carbon column, and is often used in heap leaching [33]. Oraby et al. (2020) reported the recoverability of gold from a glycine-rich solution using CIP and CIL, which were insignificantly different. However, for the CIL process, there were higher undesirable impurities that were present in the activated carbon, leading to the contamination of gold products [34]. Additionally, approximately 80% of the global production of gold was produced by the CIP process [19]. Therefore, the present study focused on using the CIP process.

In the gold recovery process from an adsorbent, incineration with a moderately high temperature was suggested as a simple method to recover gold from the activated carbon [35–37]. Nonetheless, this process consumes high energy, and the adsorbent is destroyed and cannot be reused. Therefore, in many mines and industries, chemical stripping is preferred over burning. The soluble gold species appears in the eluent as the final product through chemical desorption with acids, bases, and organic solvents [38]. Until now, the processes of the Zadra and Anglo-American Research Laboratories (AARL) have generally been utilized in numerous mines and industries, where the gold-loaded activated carbon is eluted by a strong base and sodium cyanide on a flow-bed column [39]. However, they are rather complicated processes requiring numerous techniques, equipment, and chemical reagents [40]. Thus, the batch elution of gold using diluted acids is recommended in this work due to easier operation and high efficiency. In addition, the pre-elution of impurities is also performed before gold elution to protect the final crude gold from contamination. Finally, it is necessary to adopt additional methods to yield an insoluble crude gold product, such as EW and precipitation.

This study is the first that is designed to evaluate the overall process for extracting and recovering copper and gold from WPCBs using an MSG-based lixiviant under mild conditions. Moreover, fewer studies were successful in dissolving metals under alkaline conditions using MSG. However, none have yet conducted an in-depth study and reported metal extraction under a neutral pH using MSG. The goals of this work are (1) to study the effect of solder mask layer removal from WPCBs on copper and gold leaching, (2) to assess the optimal conditions for the extraction of copper and gold using an MSG-based solution, (3) to examine downstream approaches for harvesting copper and gold products, and (4) to design a new sustainable and cost-effective process for copper and gold recycling.

2. Materials and Methods

2.1. WPCBs Preparation

The WPCBs were purchased from a junk shop in Bangkok, Thailand. Unfavorable materials, such as LED bulbs, rubbers, nuts, and wires, were physically removed, and then the WPCBs were cut into small pieces (<5 cm) using the pruning shears. Afterward, the high-speed multifunction grinder HC-500Y was used to pulverize the dried WPCBs. They were then sieved through a standard 100 mesh screen (particle size < 0.150 mm) before leaching.

2.2. Aqua-Regia Digestion and Analysis of Metal Content

To investigate the initial elemental contents of WPCBs, the powdered WPCB was digested using aqua-regia [nitric acid (HNO_3) and hydrochloric acid (HCl) at a 1:3 (v/v) ratio] at room temperature for 24 h with a solid–liquid ratio of 1:20 (w/v) [41]. After 24 h, the mixture was filtered through Whatman filter paper No. 42. The metal content of WPCBs was determined using an inductively coupled plasma optical emission spectrometer (ICP-OES), model Avio 200, PerkinElmer, USA. In this work, the calibration curves were prepared with a copper standard (Merck Millipore, Temecula, CA, USA), gold standard (Sigma-Aldrich, St. Louis, MO, USA), and multi-element quality control standard 21 (PerkinElmer, Waltham, MA, USA).

2.3. Eliminating the Solder Mask Layer of WPCBs

Before powdering WPCBs, the small pieces of WPCBs were immersed undisturbed in 10 M NaOH for 24 h to remove the solder mask layer [22]. After 24 h, the samples were cleaned by rinsing with tap water several times and eventually with distilled water (DW), before drying in an oven at 80 °C overnight or until completely dry. The initial copper and gold concentration was obtained using aqua-regia digestion according to Section 2.2.

2.4. Leaching Processes and Analytical Methods

2.4.1. The First Leaching

Ajinomoto MSG (100% monosodium L-glutamate INS No. 621) was purchased from a convenience store in Bangkok. First, 25 mL of MSG solution (pH of 7.00 and concentration 0–1.5 M) supplemented with or without H_2O_2 (0–3% (w/v)) were poured into 125 mL glass conical flasks containing 1 g WPCBs or the solder-mask-free WPCBs. Subsequently, for conditions containing ozone, 10 min of ozonation (using an ozone generator MG–1000, Thailand Juicer, Thailand, 1000 mg h^{-1} of ozone production, and 1 L m^{-1} flowrate) was provided. The leaching process was performed in an orbital shaker at 150 rpm and 30 °C for 3 h.

2.4.2. The Second Leaching

The solid residue from the optimal condition of each experiment in the first leaching was separated from the leachate by centrifugation at 4000 rpm for 10 min and was then washed twice with DW. It was dried in an oven at 80 °C overnight or until completely dry and then ground again using a pestle and mortar. A total of 1 g of the powdered solid residue was leached using 25 mL of the fresh 0–1.5 M MSG solution (pH of 7.00) supplemented with or without H_2O_2 ranging from 0–3% (w/v), or it was ozonated for 10 min. The leaching process was carried out in an orbital shaker at 150 rpm and 30 °C for 3 h.

2.4.3. Analytical Methods

After leaching, the system pH and ORP were measured by a pH meter, model SevenEasy S20-K (Mettler Toledo, Switzerland) and ORP electrode with a platinum ring indicator and S7 screw head using an Ag/AgCl reference (3 M KCl) (Mettler Toledo, InLab Redox 51343200, Switzerland), respectively. The leachates were separated from the solid residue using centrifugation at 4000 rpm for 10 min (the first leaching) and 12 min (the second leaching). Then, 4 mL of the leachate was collected independently and mixed with

4 mL aqua-regia. To avoid an excessively severe reaction, the mixture was set aside for at least 12 h before hot digestion at 105 °C for 6 h, using a digester to eliminate impurities, such as glutamate. The elemental concentration in the leachates was measured by ICP-OES Avio 200, PerkinElmer, USA and the extraction of metals was calculated following Equation (3):

$$\text{Elemental extraction efficiency }(\%) = \frac{A_2}{A_1} \times 100 \tag{3}$$

where A_1 (ppm) is the initial elemental content of WPCBs or the initial elemental concentration in the (pre-eluted) gold-loaded activated carbon, and A_2 (ppm) is the elemental content of the leachate or the elemental concentration in the solution after (pre) elution.

2.5. Copper Recovery

To accelerate the recovery process, the copper-glutamate complex had to be acidified to yield copper ions and glutamic acid. Based on the species distribution of the copper-glutamate-H_2O system reported by Perea et al. (2022) and Prasetyo et al. (2020), the copper ion mainly appears at pH 1.00 [17,18]. Thus, the pH of the first leachate was adjusted to a pH of 1.00 using concentrated sulfuric acid before the EW process (the first leachate volume was increased by ~30%). The EW was performed under room temperature using 25 × 80 × 2.0 mm aluminum and titanium plates as the cathode and anode [42], respectively. They were placed firmly at a distance of about 3.4 cm in the 100 mL beaker. Then, 80 mL of the first leachate (pH 1.00) was introduced into the system and stirred at approximately 1000 rpm by a magnetic stirrer. The samples were taken at 0.5, 1, 2, 3, 4, 5, and 6 h. The electric power was supplied by Longwei DC power supply 30 V 10 A LW-K3010D, with a voltage of 25 V. Although this voltage seemed extremely high, it provided a very low current (~20 mA measured by a multimeter). This was due to high ohmic resistance caused by the presence of a large amount of amino acid in the solution [43]. After EW, the slurry was digested by hot aqua-regia according to Section 2.4.3 to remove glutamic acid before determining the elemental concentration before and after an EW using ICP-OES Avio 200, PerkinElmer, USA and the percentage of recovery was calculated following Equation (4):

$$\text{Elemental recoverability }(\%) = \frac{B_1 - B_2}{B_1} \times 100 \tag{4}$$

where B_1 (ppm) and B_2 (ppm) are the elemental concentrations before and after processing, respectively.

2.6. Adsorption and Recovery of Gold

2.6.1. Gold Adsorption

The granular coconut-shell-based activated carbon grade HRO 6 × 12 for gold mining was purchased from Carbokarn Company Limited, Thailand. As reported by Altinkaya et al., 2020, the CIP was performed with 15 g L^{-1} of activated carbon [10]. Hence, in this experiment, 80 mL of the second leachate (gold-rich solution) was supplemented by 1.2 g of the commercial activated carbon and incubated in an orbital shaker at 150 rpm and 30 °C. The samples were taken at 2, 4, 6, 8, 10, and 12 h. Subsequently, the slurry was digested by hot aqua-regia according to Section 2.4.3 before analyzing the elemental concentration using ICP-OES Avio 200, PerkinElmer, USA. Equation (4) was also used to calculate the ability of gold adsorption.

2.6.2. Pre-Elution of Impurities

After the gold adsorption process, the gold-loaded activated carbon was harvested, washed with DW, and dried in an oven at 60 °C until completely dry. To prevent the contamination of the final crude gold powder, other impurities needed to be fully removed, especially copper, iron, magnesium, and zinc. Therefore, the pre-elution of non-targeted metals was conducted before gold elution, following Ilankoon et al. (2020) [44] with modification. The dried gold-loaded activated carbon was pre-eluted using ethylene diamine tetra-acetic acid (EDTA) at various concentrations (0.01, 0.02, 0.05, 0.10, 0.25, 0.50, and 1.00 M) at 2.5 g L^{-1}, pH 6.00, and 25 °C without shaking for 15, 30, 45, and 60 min. The solution was collected and digested with hot aqua-regia according to Section 2.4.3 for elemental content analysis using ICP-OES Avio 200, PerkinElmer, USA and the efficiency was calculated by Equation (3).

2.6.3. Gold Elution

After pre-elution, the pre-eluted gold-loaded activated carbon was collected, and it was further rinsed with DW and then dried. The gold elution process was performed following Dobrowolski et al. (2012) [45] with modification. The dried pre-eluted gold-loaded activated carbon was added into 250 mL glass conical flasks containing the eluents, which was a 1:1 ratio of DW and concentrated acids (HCl, HNO_3, and aqua-regia) at 10 g L^{-1}. The sample was incubated in an orbital shaker at 150 rpm and 30 °C for 1 h. After an hour, the sample was collected and digested according to Section 2.4.3. The elemental concentration in the solution before and after desorption was determined using ICP-OES Avio 200, PerkinElmer, USA, and calculated following Equation (3).

2.6.4. Gold EW

Firstly, the pH of the gold-containing aqua-regia solution obtained after gold elution was adjusted to 2.00 [46] using KOH (the solution volume was increased by ~13%). Then, 100 mL of the pH-adjusted gold-containing aqua regia solution was added to a 150 mL beaker for gold EW using two graphite electrodes as both cathode and anode [47,48]. Their height and diameter were 10 cm and 1 cm, respectively. They were set tightly into the system, with approximately 3.5 cm between them. The system was then stirred on a magnetic stirrer at room temperature and 1000 rpm for 30, 60, 90, 120, 150, and 180 min. It was electrically supplied by a Longwei DC power supply 30 V 10 A LW-K3010D with a voltage of 3.5 V [46] and a current of 0.60 A (measured by a multimeter) throughout the experiment. The elemental concentration in the barren solution before and after EW (the sample was harvested and digested according to Section 2.4.3) was measured using ICP-OES Avio 200, PerkinElmer, USA, and calculated using Equation (4).

2.7. Statistical Analysis

Data were analyzed statistically by one-way ANOVA with post hoc Tukey's honestly significant difference test (HSD, $p < 0.05$) to examine the differences among sample means for significance, including the elemental extractions, system pH, ORP, and absorbability. The statistical analysis was performed using the SPSS statistical software platform version 23, and the data are shown as the mean ± standard deviation (SD).

3. Results and Discussion

3.1. Elemental Composition of WPCBs

The elemental concentration of WPCBs by ICP-OES analysis is provided in Table 2. The highest contents of the metals in the WPCBs were copper, iron, and zinc, respectively, which were the base metals. However, valuable metals such as gold appeared in small quantities. After the removal of the solder mask from WPCBs by 10 M NaOH, the copper and gold concentration in solder-mask-free WPCBs were 36,654.68 ± 193.33 ppm and 23.67 ± 6.29 ppm, respectively, which is insignificantly different from that of the untreated WPCBs. This result was similar to those of Jadhav and Hocheng (2015) [26] and Jadhav et al. (2016) [22], who used 10 M NaOH to eliminate the solder mask from WPCBs. Their results showed that this process did not affect the dissolution of precious metals such as gold, silver, and palladium and had a slight impact on copper dissolution.

Table 2. ICP-OES analysis of metal content in the WPCBs used in this study.

Elements	**Concentration (ppm)**	
	Untreated WPCBs	**Solder-Mask-Free WPCBs**
Au	22.55 ± 7.11	23.67 ± 6.29
Cd	4.96 ± 3.27	0.63 ± 0.01
Co	80.61 ± 0.17	4.20 ± 0.18
Cr	330.45 ± 8.11	108.75 ± 4.10
Cu	36,905.73 ± 480.23	36,654.68 ± 193.33
Fe	18,935.41 ± 391.13	726.63 ± 15.16
Mg	786.86 ± 30.07	327.24 ± 15.27
Mn	69.79 ± 2.00	63.09 ± 4.07
Mo	1.19 ± 1.22	3.26 ± 2.85
Ni	1211.51 ± 27.38	329.84 ± 28.02
Ti	1800.41 ± 36.08	268.66 ± 8.68
V	3.32 ± 0.21	0.74 ± 0.28
Zn	7291.14 ± 168.21	7242.13 ± 21.73

3.2. Leaching of The Untreated WPCBs

3.2.1. The First Leaching

Figure 1A shows a gold leaching of less than 10% at 3 h in every condition. However, the maximum copper was dissolved (48–49%) using MSG supplemented with H_2O_2 and ozonated H_2O_2 (Figure 1B). Ozonation for 10 min seemed to play no role in the extraction of metals in this study, perhaps due to the unfavorable concentration of the ozone for metal oxidation. The copper was oxidized first by an oxidizer, specifically, H_2O_2. According to Perea et al., 2022, the copper ions were then chelated with MSG to form copper-glutamate species (Equations (5) and (6)) [18].

$$Cu^{2+}\,(aq) + C_5H_8NO_4Na\,(aq) \leftrightarrow Cu\big(C_5H_8NO_4\big)^{+}\,(aq) + Na^{+}\,(aq) \tag{5}$$

$$Cu^{2+}\,(aq) + 2(C_5H_8NO_4Na)\,(aq) \leftrightarrow Cu\big(C_5H_8NO_4\big)_2\,(aq) + 2Na^{+}\,(aq) \tag{6}$$

Figure 1C demonstrates that the addition of oxidizing agent(s) resulted in an obvious decrease in the system pH due to the acidic nature of oxidizing agents. In the MSG-rich conditions, the system pH was 7.29 and remained between 6.97 and 7.22 regardless of the presence of the oxidizing agent(s). This was mainly due to three main reasons: (1) the OH^- production according to Equations (1) and (2), (2) the generation of sodium ions, which are a byproduct from the metal-MSG complexation (Equations (5) and (6)), and (3) the dissolution of alkaline metals from WPCBs.

Unsurprisingly, in comparison with DW without the oxidizer, increased ORP results were observed in H_2O_2-rich DW (Figure 1D), whereas the 10 min ozonation was responsible for a slight decrease in the ORP. This was probably because of the generation of reductive species after adding the ozone. The presence of H_2O_2 and ozonated H_2O_2 in the solution could be attributed to the highest ORP values, indicating the highest metal oxidation capacity. Moreover, the ORP of the DW-based conditions was significantly higher than those of the MSG-based conditions, possibly because of the interference of amino acids and the generation of reductive species [49].

Previous studies revealed that gold could be significantly leached from WPCBs containing small amounts of base metal, particularly copper, using an amino acid-based solution. Thus, to achieve the highest gold leaching, the second leaching by treating WPCB residues with a fresh lixiviant was proposed [9,20]. This circumstance is because the E^0 value of gold (Au^+/Au = 1.69 V vs. standard hydrogen electrode (SHE) is higher than that of copper (Cu^{2+}/Cu = 0.34 V vs. SHE), as mentioned above. In this study, the second leaching was performed, and solid residues were obtained by subjecting WPCB to 1 M MSG supplemented with 1% (w/v) H_2O_2.

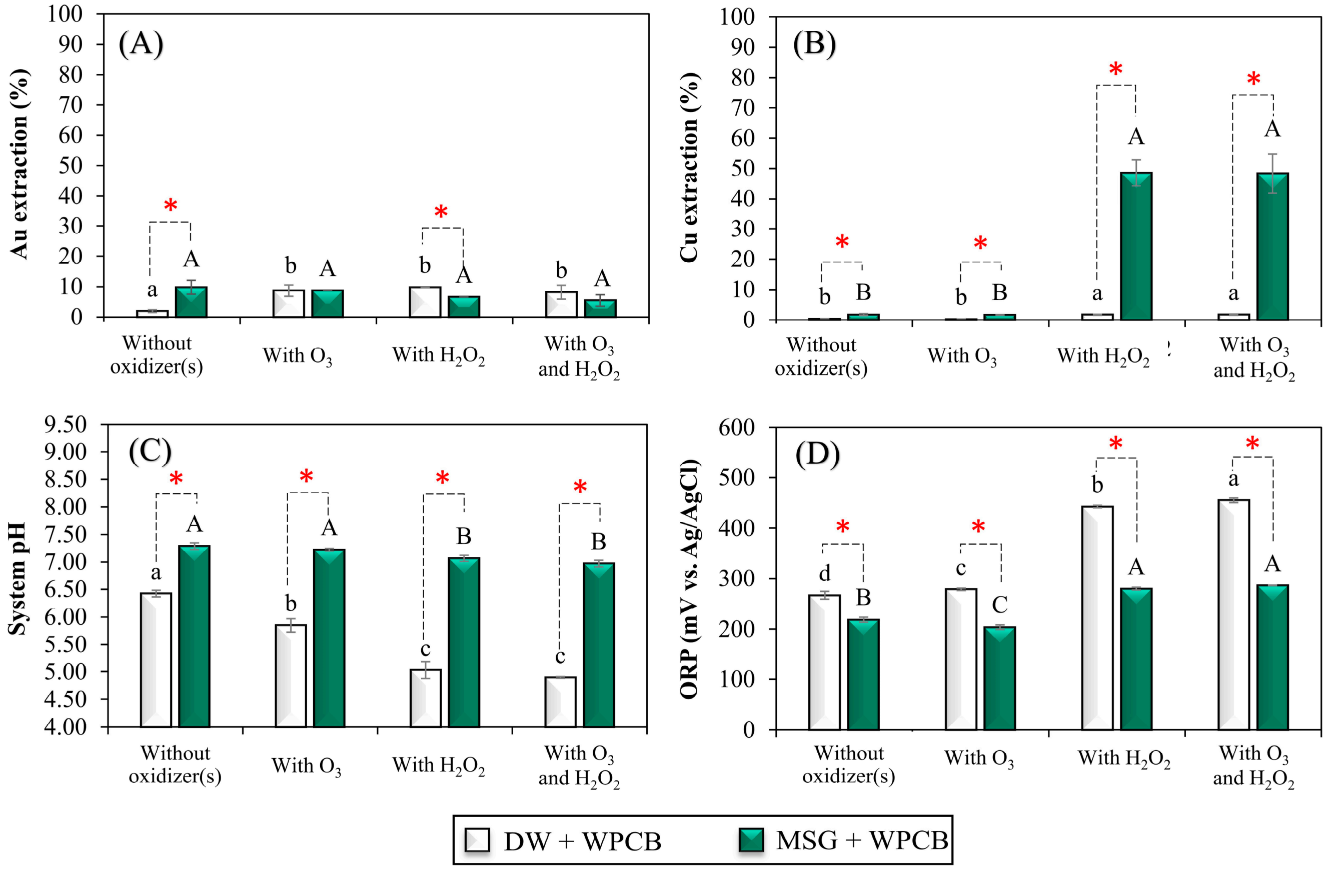

Figure 1. Gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from WPCBs using 1 M MSG with and without 1% (w/v) H_2O_2 and 10 min ozonation (O_3 concentration = 167 mg L^{-1}) at 4% pulp density for 3 h; different lowercase and capital letters on the bars indicate significant differences ($p < 0.05$) of DW+WPCB and MSG+WPCB for each treatment, respectively, while the asterisk (*) designates the significant differences ($p < 0.05$) under the same treatment; data are presented as the mean ± SD (n = 3).

3.2.2. The Second Leaching

At 3 h, the greatest extractions of gold (20%) were obtained using MSG supplemented with H_2O_2 and MSG supplemented with O_3. Nevertheless, the combination of H_2O_2 and O_3 led to a decrease in gold extractability (Figure 2A), due most likely to the MSG oxidation by excess oxidizers, especially $^{\bullet}OH$. Theoretically, the oxidizing agent played an important role in oxidizing the gold from WPCBs. The gold ions were then complexed with amino acids under neutral conditions [10]. The reaction between gold and MSG is shown in Equation (7).

$$Au^{+}(aq) + 2C_5H_8NO_4Na\ (aq) \rightarrow Au(C_5H_8NO_4)_2^{-} + 2Na^{+}\ (aq) \tag{7}$$

Figure 2B shows that the use of MSG supplemented with H_2O_2 led to the highest copper extraction potential of approximately 77% from the WPCB residue. Similar to gold, the reduced leaching of copper was found in the mixture of MSG, H_2O_2, and O_3, possibly due to the MSG oxidation.

The highest system pH (7.36) appeared when using MSG supplemented with H_2O_2 within 3 h under neutral conditions (Figure 2C), which corresponded to the maximum dissolution of metals. In contrast, MSG supplemented with H_2O_2 condition was influential in the lowest ORP level (121 mV vs. Ag/AgCl) (Figure 2D), likely due to the generation of reductive species.

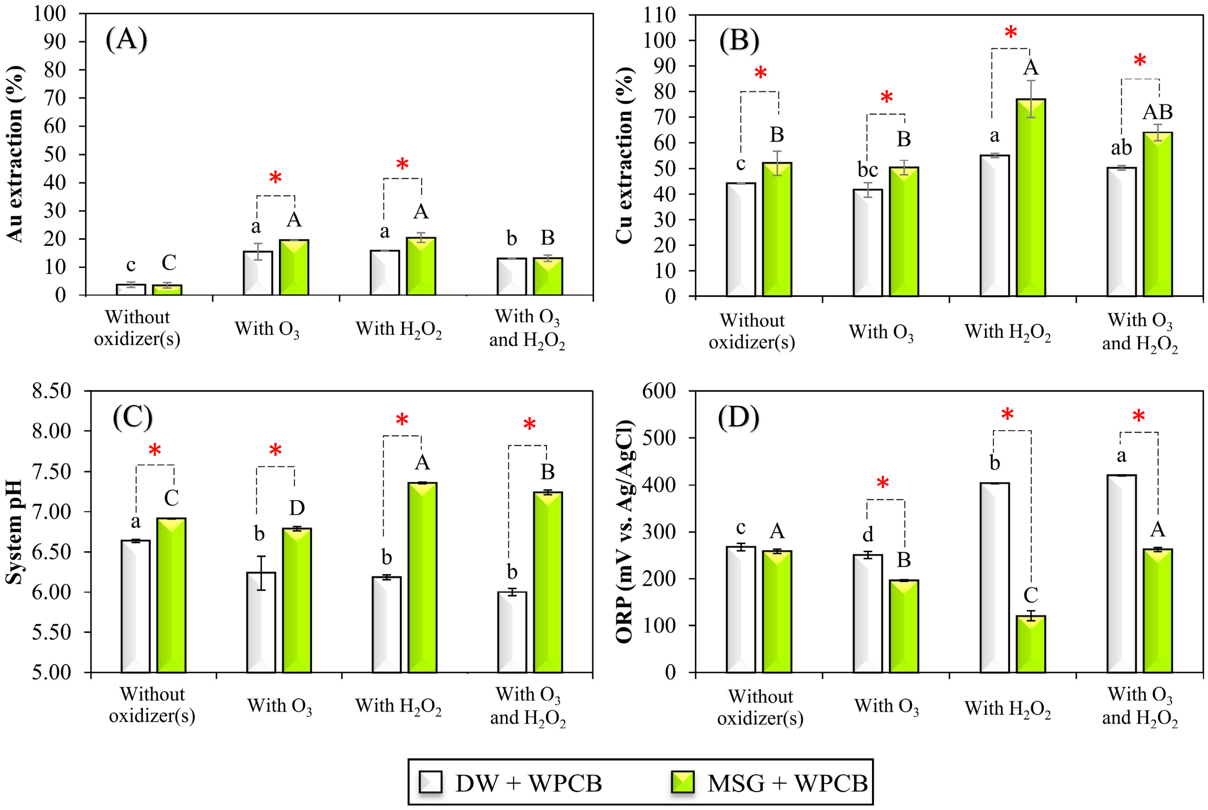

Figure 2. Gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from the WPCB residue (obtained from the first leaching using 1 M MSG supplemented with 1% (w/v) H_2O_2 at 4% pulp density) using the fresh 1 M MSG with and without 1% (w/v) H_2O_2 and 10 min ozonation (O_3 concentration = 167 mg L^{-1}) at 4% pulp density for 3 h; different lowercase and capital letters on the bars indicate significant differences ($p < 0.05$) of DW + WPCB and MSG + WPCB for each treatment, respectively, while the asterisk (*) designates the significant differences ($p < 0.05$) under the same treatment; data are presented as the mean ± SD (n = 3).

3.3. Leaching of The Solder-Mask-Free WPCBs

3.3.1. The First Leaching

As can be seen in Figure 3A,B, the removal of the solder mask layer from WPCBs contributed to a considerable increase in copper leaching. However, the gold extraction efficiency did not improve (there was no condition where the extraction of gold exceeded 10%). This behavior confirmed that gold extraction from copper-rich samples is highly difficult. At 3 h, the highest copper extractability (74%) was achieved using MSG supplemented with H_2O_2, probably due to the higher penetrability of the leaching agent [22]. Hence, this lixiviant was selected to prepare the solid residue for the second leaching.

The system pH of the DW-based solution and MSG-based solution without an oxidizer were 10.45 and 8.17, respectively (Figure 3C). Similar to Figure 1C, the addition of oxidizer(s) allowed for a significant reduction in the system pH. Interestingly, the system pH that used MSG-based solutions to leach the metals from solder-mask-free WPCBs was higher than that of the WPCBs, likely because of the greater dissolution of metals and OH^- production (Equations (1) and (2)), especially copper.

The maximum ORP was attained from the ozonated DW-H_2O_2 and ozonated MSG-H_2O_2 systems, approximately 256 mV vs. Ag/AgCl and 234 mV vs. Ag/AgCl, respectively (Figure 3D). However, in these conditions, the copper extractions were lower than that of the H_2O_2 supplementation without O_3 (Figure 3B). This observation confirmed the oxidation of MSG in an environment with excess oxidizers.

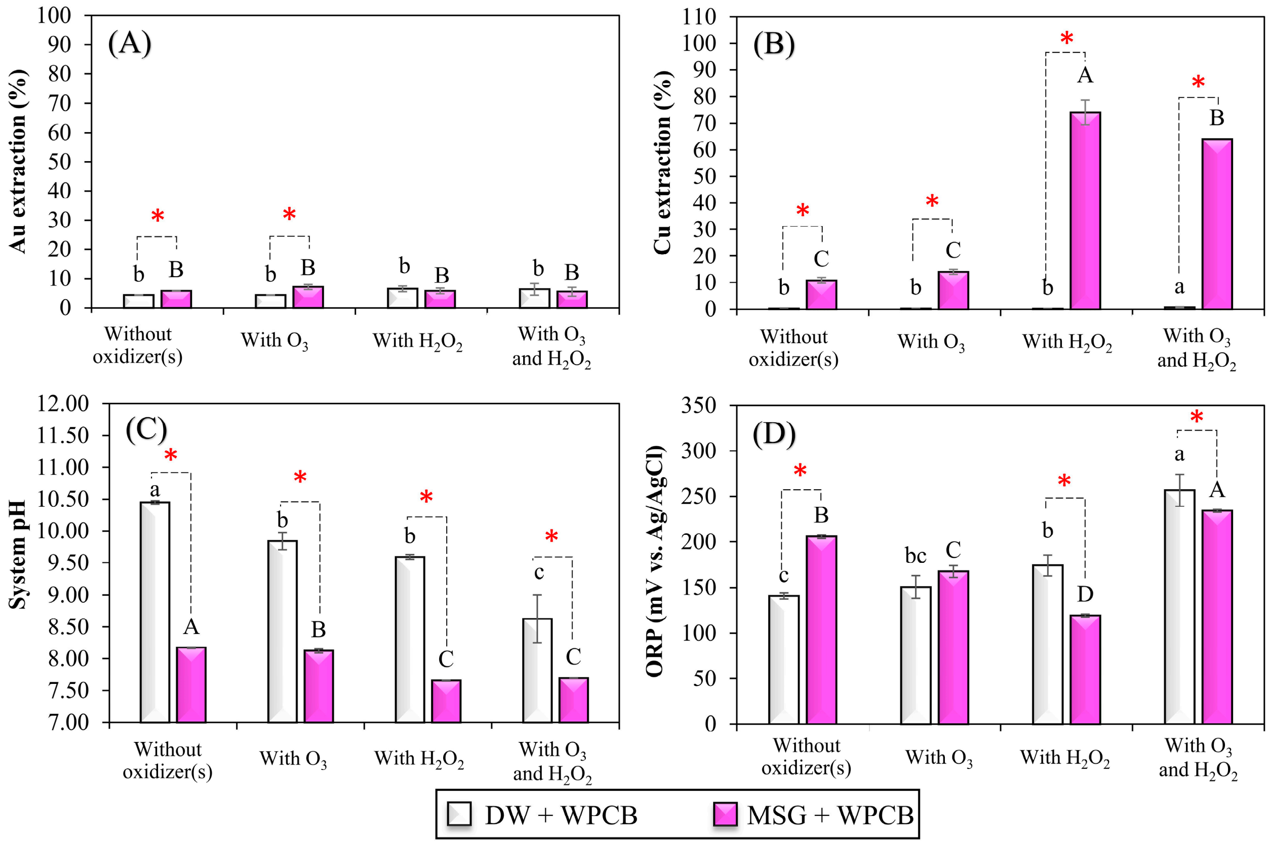

Figure 3. Gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from solder-mask-free WPCBs using 1 M MSG with and without 1% (*w*/*v*) H_2O_2 and 10 min ozonation (O_3 concentration = 167 mg L^{-1}) at 4% pulp density for 3 h; different lowercase and capital letters on the bars indicate significant differences ($p < 0.05$) of DW + WPCB and MSG + WPCB for each treatment, respectively, while the asterisk (*) designates significant differences ($p < 0.05$) under the same treatment; data are presented as the mean ± SD (n = 3).

3.3.2. The Second Leaching

In the second leaching, the gold was leached about 4% by MSG without an oxidizer within 3 h from the solder-mask-free WPCB residue obtained from the first leaching using 1 M MSG supplemented with 1% (*w*/*v*) H_2O_2 (Figure 4A). The gold leaching was enhanced enormously from 81% to 87% by the addition of oxidizer(s) to MSG, and there was no significant difference between using O_3, H_2O_2, and ozonated H_2O_2, likely due to most of the copper being leached in the first leaching. However, for practical applications, the ozonation of MSG was more difficult to manipulate, was pricier, and had more power consumption than the addition of H_2O_2. Therefore, the H_2O_2-supplemented MSG was chosen as the suitable lixiviant to extract the gold in the second leaching for optimization in subsequent experiments.

According to Figure 4B, the copper was dissolved to 24% from the solid residue in the second leaching using the MSG solution without an oxidizer. It was maximized to 38% by using MSG supplemented with H_2O_2.

The addition of oxidizer(s) resulted in a decrease in the system pH (Figure 4C) and an increase in ORP (Figure 4D). The highest drop in the system pH (6.50) was presented using ozonated MSG supplemented with H_2O_2. On the contrary, the maximum ORP value of the MSG-based systems (317 mV vs. Ag/AgCl) was observed in this condition, which was not significantly different from using MSG in combination with H_2O_2 (303 mV vs. Ag/AgCl).

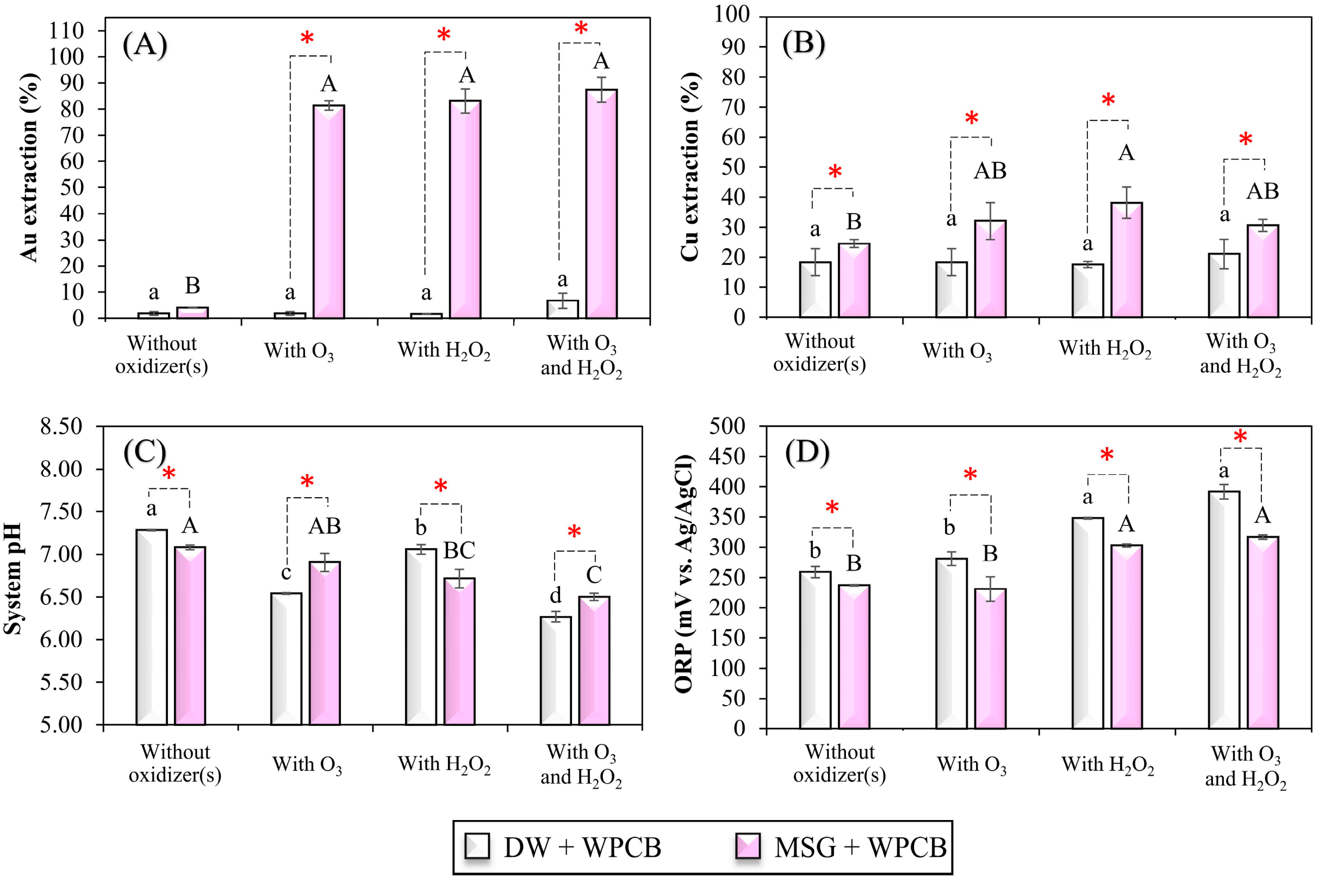

Figure 4. Gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from the solder-mask-free WPCB residue (obtained from the first leaching using 1 M MSG supplemented with 1% (*w*/*v*) H_2O_2 at 4% pulp density) using the fresh 1 M MSG with and without 1% (*w*/*v*) H_2O_2 and 10 min ozonation (O_3 concentration = 167 mg L^{-1}) at 4% pulp density for 3 h; different lowercase and capital letters on the bars indicate significant differences ($p < 0.05$) of DW + WPCB and MSG + WPCB for each treatment, respectively, while the asterisk (*) designates the significant differences ($p < 0.05$) under the same treatment; data are presented as the mean ± SD (n = 3).

3.4. MSG Concentration

3.4.1. The First Leaching

The extractions of gold and copper using 1% (*w*/*v*) H_2O_2 in combination with MSG at different concentrations (0–1.5 M) are shown in Figure 5A,B. The results indicate that the highest extraction levels of gold and copper were obtained using 0.75 M MSG supplemented with 1% (*w*/*v*) H_2O_2 at 7% and 82%, respectively, for 3 h under neutral conditions. Lower MSG concentrations (0–0.5 M) were insufficient for the maximum extractions of gold and copper from solder-mask-free WPCBs, while excessive MSG concentrations (1–1.5 M) led to a steric effect [50]. These results were similar to Oraby et al. (2017), who studied the dissolution of gold and copper from gold-copper ores and concentrated them using glycine. Their result indicated that the use of excessively high glycine concentration (10 g L^{-1}) provided a reduction in gold leaching compared to their optimal glycine concentration (5 g L^{-1}) [51].

At 3 h, the system pH of the DW-H_2O_2 condition (0 M MSG) was about 9.22 (Figure 5C). The addition of MSG was responsible for the considerable reduction in the system pH to 6.96. The system pH increased continually in conjunction with the enhancement of the MSG concentration, perhaps due to the presence of sodium ions, the release of alkaline metals, and OH^- generation (Equations (1) and (2)). The system pH was insignificantly different when the MSG concentration was above 0.75 M, which was related to the highest gold and copper extraction abilities (Figure 5A,B).

The ORP of the DW-H_2O_2 condition was approximately 181 mV vs. Ag/AgCl (Figure 5D). Small amounts of MSG seemed to influence an increase in ORP. However, the ORP continued to decrease while increasing the concentration of MSG, likely due to the higher concentration of glutamate.

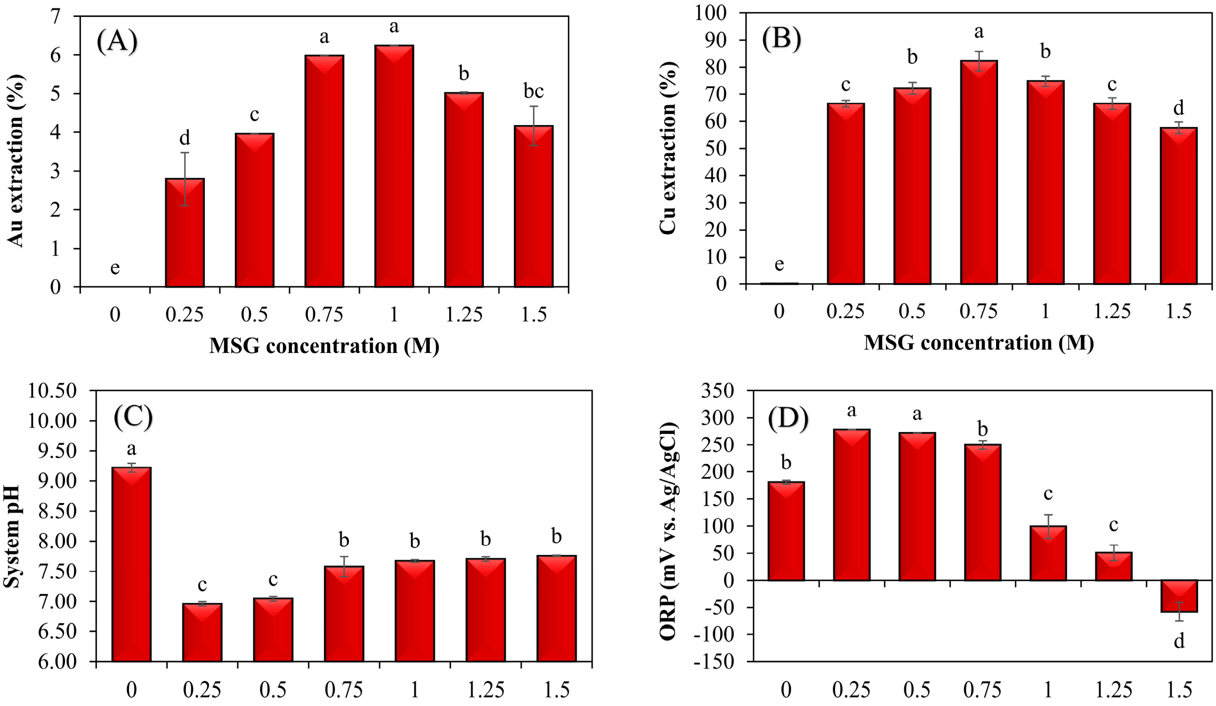

Figure 5. Effect of MSG concentration on gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from solder-mask-free WPCBs using 1% (*w*/*v*) H_2O_2 as an oxidizer at 4% (*w*/*v*) pulp density for 3 h; different lowercase letters on the bars indicate significant differences ($p < 0.05$); data are presented as the mean ± SD (n = 3).

3.4.2. The Second Leaching

The solder-mask-free WPCB residue was prepared using the optimal lixiviant from the previous result (0.75 M MSG supplemented with 1% (*w*/*v*) H_2O_2), and it was re-leached by a fresh lixiviant (MSG supplemented with H_2O_2) at different MSG concentrations (0–1.5 M). Figure 6A,B show that the greatest extractions of gold (81%) and copper (36%) were achieved by using 1 M MSG coupled with 1% (*w*/*v*) H_2O_2. Similar to Figure 5A,B, the metal extractions were insignificantly different or reduced with the increasing MSG concentration.

At 3 h, the system pH decreased continually with the increasing MSG concentration (Figure 6C). The lowest system pH (6.42) was attained from the use of 0.5 M MSG. A higher MSG concentration from this point led to an increase in the system pH and stabilized at 6.81–6.90 when the MSG concentration was over or equal to 1.25 M.

Figure 6D demonstrates that the enhancement of MSG proportions resulted from the reduction in ORP, likely owing to higher contents of glutamate and reductive species.

From these reported results (Figures 5 and 6), 0.75 M MSG and 1 M MSG were chosen as the optimal MSG concentration for copper extraction in the first leaching and gold extraction in the second leaching and, respectively, in subsequent trials.

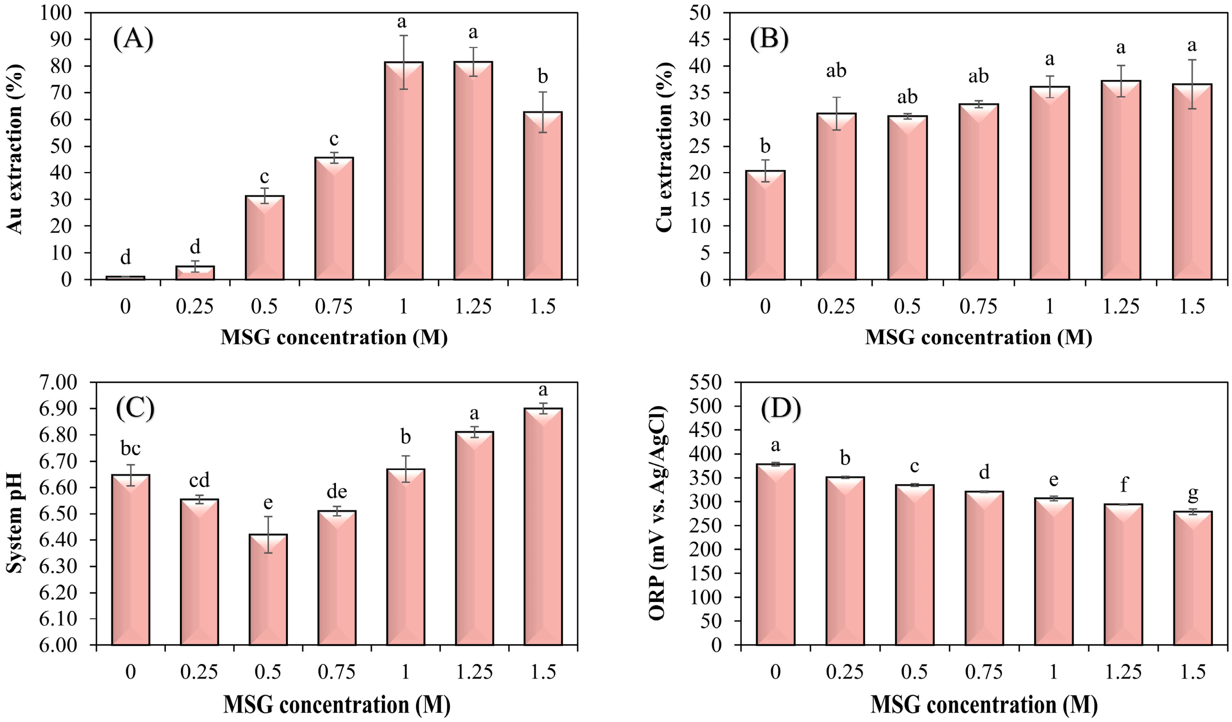

Figure 6. Effect of MSG concentration on gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from the solder-mask-free WPCB residue (obtained from the first leaching using 0.75 M MSG supplemented with 1% (*w*/*v*) H_2O_2 at 4% pulp density) using a fresh 1% (*w*/*v*) H_2O_2 as an oxidizer at 4% (*w*/*v*) pulp density for 3 h; different lowercase letters on the bars indicate significant differences ($p < 0.05$); data are presented as the mean ± SD (n = 3).

3.5. H_2O_2 Concentration

3.5.1. The First Leaching

Figure 7A displays the effects of the H_2O_2 concentration (0–3% (*w*/*v*)) on the leaching of gold from solder-mask-free WPCBs. Almost no gold extraction was observed when the concentration of H_2O_2 was below 1%. The gold was dissolved at a maximum of 10% using 1% and 1.5% (*w*/*v*) H_2O_2 in combination with 0.75 M MSG. However, no gold could be detected in the leachate when H_2O_2 above 1.5% (*w*/*v*) was used, which is because of the oxidization of MSG by excessive H_2O_2. Likewise, the decreasing extraction of gold with the increasing oxidizer concentration was reported by Oraby et al. (2020). They evaluated the extraction of gold from oxide ores using alkaline glycine in the presence of permanganate ions. Their result demonstrated that gold extraction after 48 h was reduced by approximately 8.5% when the permanganate concentration was enhanced from 1 to 2 g L^{-1}, indicating that the glycine was oxidized under the condition with excess permanganate ions [34].

The copper leaching efficiency increased significantly from 18% to 84%, as the concentration of H_2O_2 rose from 0 to 0.1% (*w*/*v*) (Figure 7B). The highest copper extraction level (93%) was obtained when using 0.75 M MSG supplemented with 0.5% (*w*/*v*) H_2O_2. The enhancement of the concentration of H_2O_2 over 0.5% (*w*/*v*) led to a significant decrease in copper leaching. Thus, the concentration of H_2O_2 at 0.5% (*w*/*v*) was chosen for further investigation.

The system pH (Figure 7C) and the ORP (Figure 7D) were directly and inversely proportional to the increase in the concentration of H_2O_2, respectively. The system pH decreased dramatically from 8.18 to 6.63, possibly because of the acidic nature of H_2O_2. The ORP increased greatly, from 93 mV to 307 mV vs. Ag/AgCl, indicating the high oxidation of the metals. Although the system pH and ORP changed significantly from an optimal point (0.5% (*w*/*v*) H_2O_2), the extraction of metals, especially copper, was reduced. This behavior confirmed the oxidation of MSG under excess H_2O_2 conditions.

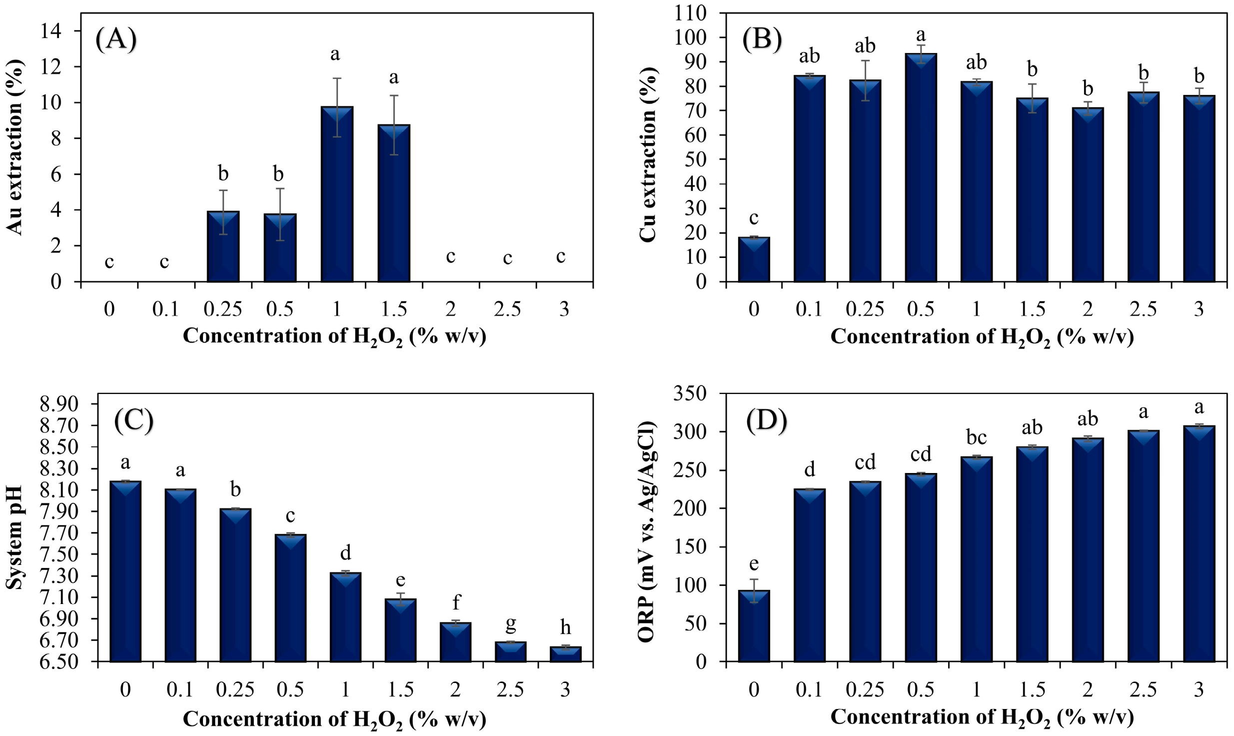

Figure 7. Effect of H_2O_2 concentration on gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from solder-mask-free WPCBs using 0.75 M MSG as a chelator at 4% (*w*/*v*) pulp density for 3 h; different lowercase letters on the bars indicate significant differences ($p < 0.05$); data are presented as the mean ± SD (n = 3).

3.5.2. The Second Leaching

In this study, the solder-mask-free WPCB residue was obtained from the first leaching using 0.75 M MSG supplemented with 0.5% (*w*/*v*) H_2O_2. Figure 8A shows the effect of the concentration of H_2O_2, ranging from 0% to 3%, on the leaching of gold when the concentrations of MSG were kept constant at 1 M. The gold extraction increased rapidly and reached the maximum value of 86% within 3 h at the concentration of H_2O_2, ranging from 0.25% to 2.5% (*w*/*v*). Nevertheless, the increasing concentration of H_2O_2 from 2.5% to 3% (*w*/*v*) caused a 20% decrease in the leaching potential of gold, likely due to the denaturation of glutamate in the solution. Hence, further second leaching was performed using 1 M MSG supplemented with 0.25% (*w*/*v*) H_2O_2.

As presented in Figure 8B, the enhancement of H_2O_2 concentration from 0% to 3% (*w*/*v*) did not lead to the higher leaching efficiency of copper from the solder-mask-free WPCB residue (about 7% remaining), indicating the limitation of copper dissolution in this system.

The system pH began to decrease when the concentration of H_2O_2 was higher than 0.5% (*w*/*v*) (Figure 8C). Moreover, the higher concentration of H_2O_2 increased the ORP continually (Figure 8D). However, as described above, the excess H_2O_2 concentration (>3% (*w*/*v*)) reduced gold leaching due to the denaturation of MSG.

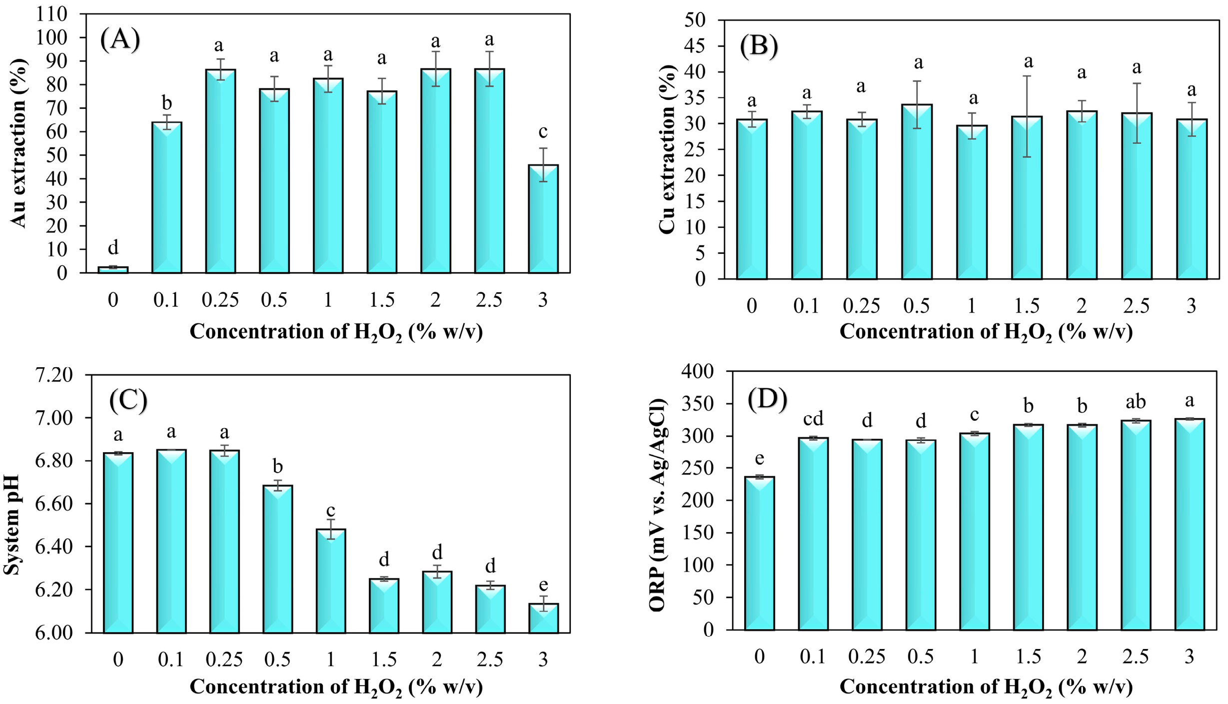

Figure 8. Effect of H_2O_2 concentration on gold leaching (**A**), copper leaching (**B**), system pH (**C**), and ORP (**D**) from the solder-mask-free WPCB residue (obtained from the first leaching using 0.75 M MSG supplemented with 0.5% (*w*/*v*) H_2O_2 at 4% pulp density) using 1 M MSG as a chelator at 4% (*w*/*v*) pulp density for 3 h; different lowercase letters on the bars indicate significant differences ($p < 0.05$); data are presented as the mean ± SD (n = 3).

3.6. Impurities in the Leachates

Based on the previous results, the optimal conditions for the first leaching and the second leaching were different. To maximize the copper leaching of the first leaching, the process was conducted under neutral conditions (30 °C, 150 rpm, and initial pH of lixiviant of 7.00) at 4% pulp density for 3 h using 0.75 M MSG coupled with 0.5% (*w*/*v*) H_2O_2. Figure 9A shows the impurities in the first leachate. It was found that the MSG-H_2O_2 system was highly suitable for the extraction of copper because almost 100% of the copper was dissolved from the solder-mask-free WPCBs with low contamination of other unfavorable metals. Li et al. (2022) revealed that the high copper concentration in the solution provided a possibility of direct copper EW [20]. In addition, the copper concentration of the first leachate of this study was ~ 35 g L^{-1}. As reported by Sun et al. (2015), copper concentration in the leachate beyond 20 g L^{-1} was suitable for direct industrial copper EW [52]. This information led to the direct recovery of copper from the first leachate without the purification and separation process, which was cost- and time-saving. This is especially useful in the development of the industrial level for maximum economic benefit. Conversely, the optimal condition for gold leaching in the second stage was achieved using 1 M MSG supplemented with 0.25% (*w*/*v*) H_2O_2 at 4% pulp density for 3 h. However, not only gold was extracted in this stage, but also other metals, especially zinc, copper, iron, and magnesium (Figure 9B). Hence, further purification was necessary before the gold recovery process.

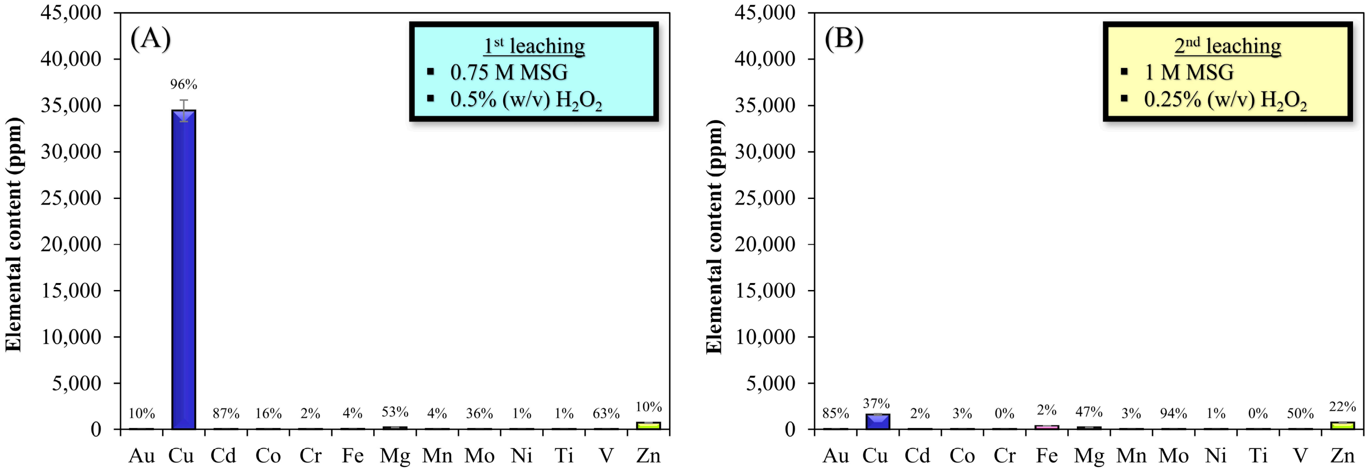

Figure 9. The extracted elements under optimal conditions from solder-mask-free WPCBs (**A**) and solid residues (**B**) in the leachate of the first leaching (0.75 M MSG, 0.5% (*w*/*v*) H_2O_2) and the second leaching (1 M MSG, 0.25% (*w*/*v*) H_2O_2), respectively; the percentages on the bar represent the extractions (%); data are presented as the mean ± SD (n = 3).

3.7. Copper Recovery

The copper was recovered from the first leachate (copper-rich solution) using EW. To accelerate the copper electrorefining, the adjustment of pH is necessary. In this study, the pH of the first leachate was adjusted to 1.00 using concentrated sulfuric acid. The selective recovery of copper ranged from 98 to 100% within 5 to 6 h, alongside the low contamination of other metals such as iron, magnesium, and zinc (Figure 10). However, to maximize the economic benefits, we suggested that process optimization is a key point that needs to be studied in the future, such as voltage, current density, and the distance of the cathode and anode.

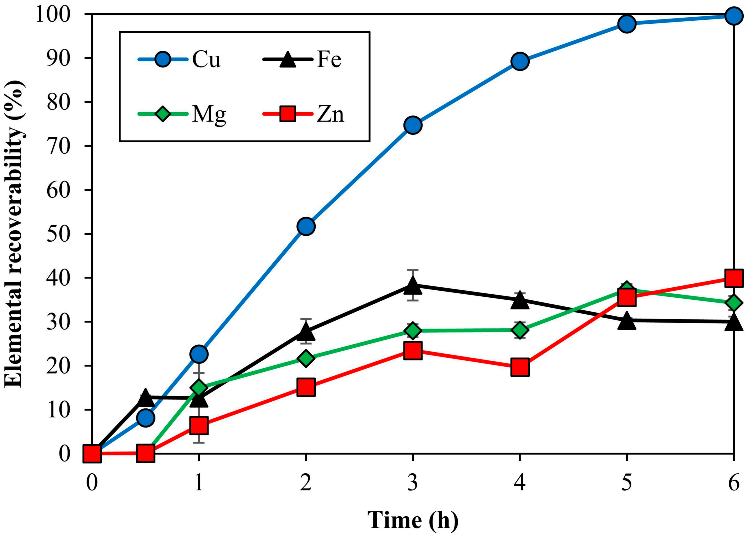

Figure 10. The recoverability of metals in the first leachate using electrowinning (EW); data are presented as the mean ± SD (n = 3).

3.8. Gold Recovery

3.8.1. Gold Adsorption

The nearly complete gold adsorption efficiency (98%) was achieved within 4 h from the second leachate (gold-rich solution containing ~18 ppm gold) using 15 g L^{-1} commercial activated carbon (Figure 11) and less than 0.4 ppm of residual soluble gold in the solution after adsorption. This result was similar to that of Altinkaya et al. (2020), who reported a 100% recovery of gold at 4 h from the gold-rich glycine solution using commercial activated carbon (RO 3515, Norit, technical grade) [10]. Additionally, the sodium ion from MSG in the second leachate could facilitate gold glutamate-complex adsorption. Following Davidson (1974) and McDougall et al. (1980), the adsorption of gold cyanide complex onto the surface-active sites of the activated carbon was unable to occur without stabilizing cations such as calcium, magnesium, and sodium ions. These stabilizing cations could electrostatically attract the negatively charged gold cyanide complex. The adsorption strength was strongly dependent on the presence of stabilizing cations in the complex, as follows: calcium ion > magnesium ion > hydrogen ion > lithium ion > sodium ion > potassium ion [53,54].

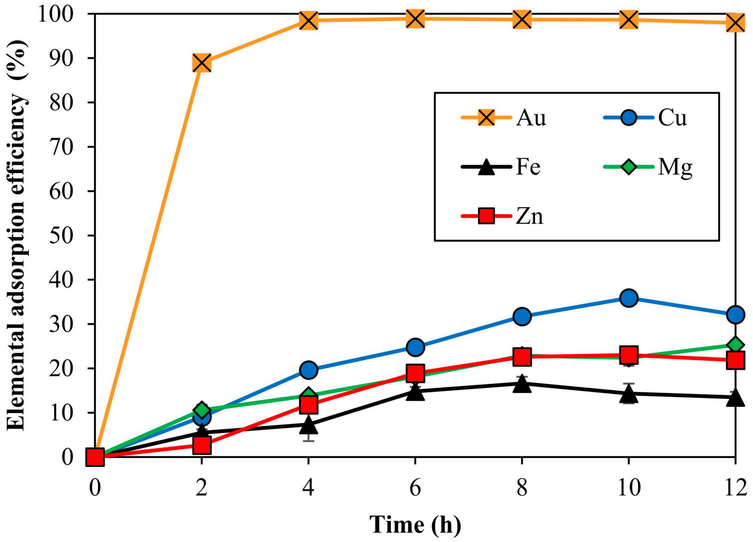

Figure 11. The elemental adsorption efficiency from the second leachate using activated carbon; data are presented as the mean ± SD (n = 3).

3.8.2. Pre-Elution of Impurities

The removal of unfavorable metal contaminants, especially copper, iron, magnesium, and zinc, present on 1 g gold-loaded activated carbon (~14 µg gold) using 0.05 M EDTA was attempted. The result (Figure 12A) indicated that the zinc was completely desorbed within the half-hour without gold dissolution. The magnesium started releasing at 30 min and continued to increase to 18% at the end of the experiment. The maximum elution of copper (23%) and iron (33%) were observed at 30 min and 15 min, respectively, and then leveled off or remained constant, indicating the limitation of the metal elution efficiencies using this condition. However, the residual of non-targeted metals that remained on the gold-loaded activated carbon was still high. Hence, the elution of unwanted metals using EDTA was conducted again for 30 min by varying the EDTA concentration, and the result is shown in Figure 12B. It was found that the highest elution of copper (77%), iron (53%), magnesium (6%), and zinc (100%) were attained using EDTA at concentrations of 0.05 M, 0.50 M, 0.25 M, and 0.01 M, respectively. Interestingly, gold was not eluted in all the EDTA concentrations tested. The reason behind this may be due to the preferentially strong chelation between EDTA and many base metals and their stability constants [44]. Overall, the pre-elution of impurities on the gold-loaded activated carbon in 0.05 M of EDTA solution for 30 min seemed to be an optimal condition in this work, as most of the contaminants were removed, particularly copper and zinc.

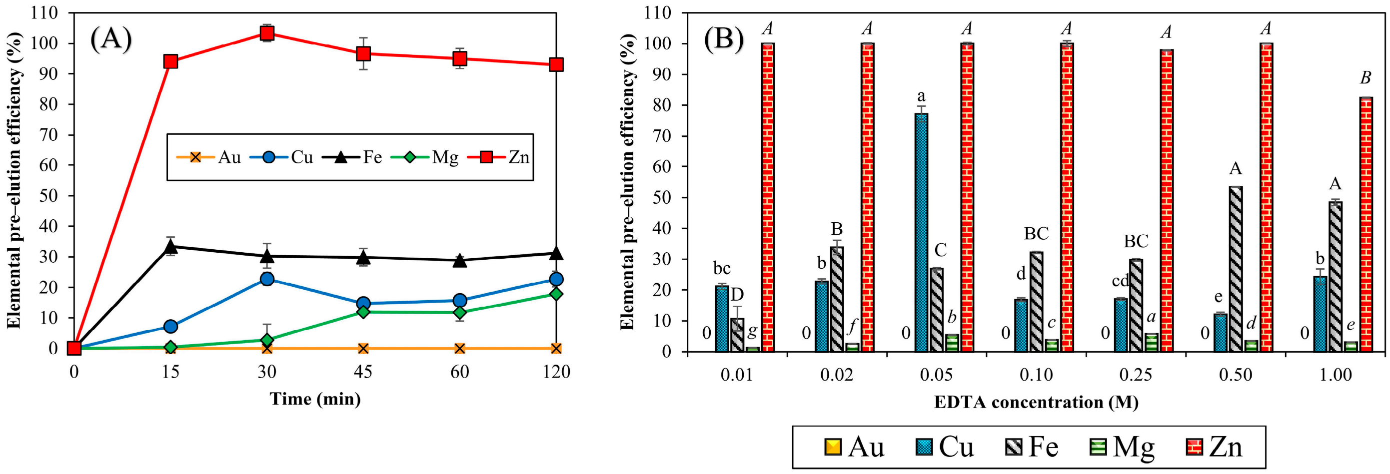

Figure 12. The pre-elution efficiency of metals from gold-loaded activated carbon using 0.02 M EDTA at various times (**A**); the effect of EDTA concentration on the pre-elution efficiency of metals from gold-loaded activated carbon at 30 min (**B**); different lowercase letters, capital letters, italic lowercase letters, and italic capital letters on the bars indicate significant differences ($p < 0.05$) in the pre-elution efficiencies of copper, iron, magnesium, and zinc for each treatment, respectively; data are presented as the mean ± SD (n = 3).

3.8.3. Gold Elution

In the gold desorption process, the mixtures of DW and several concentrated acids (HCl, HNO_3, and aqua-regia) at a ratio of 1:1 (v/v) were used as the eluents to desorb gold from the pre-eluted gold-loaded activated carbon which was obtained after 4 h adsorption and 30 min pre-elution. From Figure 13, the results demonstrate that using diluted HCl could completely desorb the unfavorable base metals (copper, iron, and magnesium) within an hour. However, only 4% gold was detected in the HCl eluent. This result was similar to that of Adams and Fleming (1989), who employed 0.1 M HCl to wash the gold-loaded activated carbon and reported that no gold was detected in the solution after 24 h [55]. Tu et al. (2011) also found that HCl alone was not able to elute valuable metals such as gold, platinum, and palladium from the modified and activated carbon [56]. In comparison, under the same conditions, lower concentrations of contaminants were observed in the diluted HNO_3 and the diluted aqua-regia. The gold on the pre-eluted gold-loaded activated carbon could be released at approximately 88% using the diluted HNO_3. Moreover, gold was successfully desorbed by the diluted aqua-regia due to the well-known synergy of HCl and HNO_3. Briefly, the gold was initially oxidized by HNO_3, and this allowed for the opportunity of the potential desorption of the gold complex from activated carbon and ultimately led to the formation of auric chloride $\left(AuCl_4^-\right)$ in the solution [57]. However, all types of acids used in this work could also highly desorb the other unwanted metals, especially iron, copper, and magnesium. Consequently, both eluting agents and conditions, as key factors that can accomplish the highly selective gold desorption with low or zero contamination, need to be further studied.

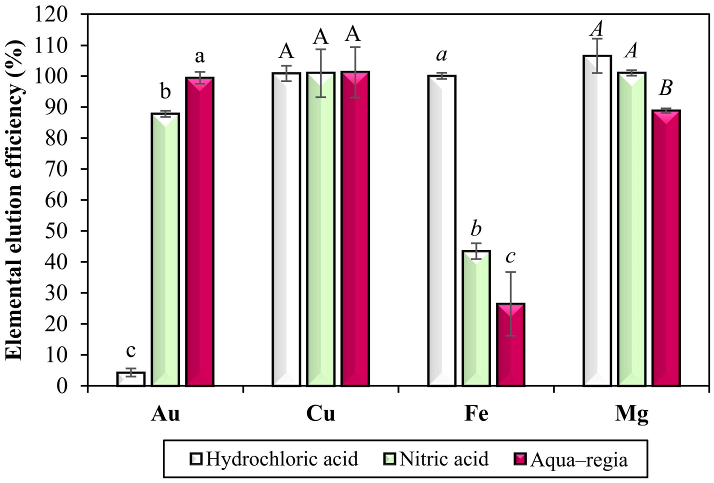

Figure 13. The elution efficiency of metals from the pre-eluted gold-loaded activated carbon; the lowercase letters, the capital letters, the italic lowercase letters, and italc capital letters on the bars indicate significant differences ($p < 0.05$) in the elution efficiencies of gold, copper, iron, and magnesium for each treatment, respectively; data are presented as the mean ± SD (n = 3).

3.8.4. Recovery of Gold by Electrowinning (EW)

To examine the performance of gold recovery by EW, the pH of the aqua-regia containing the gold solution was first adjusted to 2.00. This pH was proper for gold EW, as reported by Lee et al. (1997) [46]. Furthermore, at this pH, the graphite electrode could be slightly corroded by the oxidizing acid, namely HNO_3, which was observed at the end of the process. After supplying 3.5 V to the system, gold was successfully electrowon within 90 min and remained constant during the electrolytic period (Figure 14). However, other unfavorable metals such as copper, iron, and magnesium also deposited on the cathodic surface at approximately 78%, 46%, and 32%, respectively. When considering the purity of the final gold product, it is preferable to prolong the electrodeposition to 120 min since copper and magnesium became detached from the cathodic surface, resulting in an approximately 26% and 32% reduction in recoverability, respectively. This behavior was likely due to the improper operative condition conducted in this work, such as the pH of the solution and the type of cathode or anode used. Nevertheless, copper and magnesium were reclaimed again after 120 min, leading to higher contamination of gold. Thus, 120 min was selected as the optimal time for EW in this study.

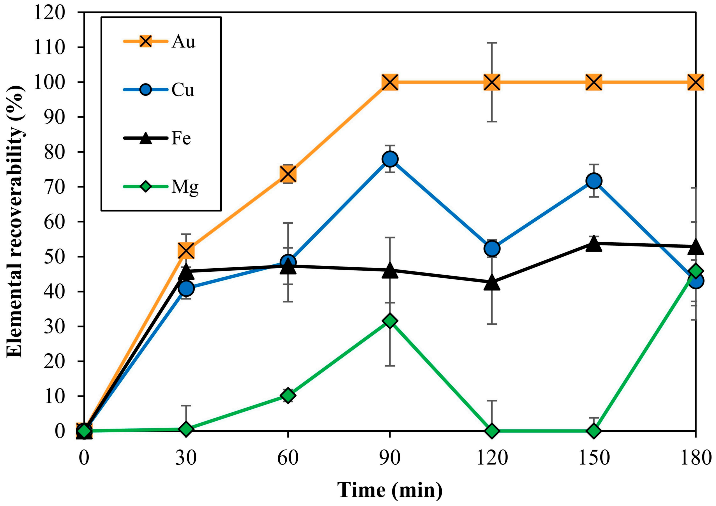

Figure 14. The elemental recovery by electrowinning (EW) from the diluted aqua-regia containing gold (initial pH 2.00); data are presented as the mean ± SD (n = 3).

3.9. Overall Process for Recovery of Copper and Gold from Waste Printed Circuit Board by Monosodium Glutamate Supplemented with Hydrogen Peroxide

Overall, the hydrometallurgical method employed in this study for the extractions of copper and gold is summarized and presented in Figure 15. The solder mask layer needed to be removed from the WPCB sample before the leaching process to improve the metal extractions. The proposed MSG-based leaching approach under mild conditions (30°C, 150 rpm, and initial pH of lixiviant of 7.00) comprised two main steps: (1) the first leaching used 0.75 M MSG and 0.5% (*w*/*v*) H_2O_2 for the extraction of the base metal, especially copper, from the solder-mask-free WPCBs and (2) the second leaching by 1 M MSG and 0.25% (*w*/*v*) H_2O_2 helped to dissolve the valuable metal, particularly gold, from the first residue. Subsequently, the separation and recovery of copper and gold were performed. The copper could be harvested directly from the first leachate using the EW due to low contaminations of other metals. Many processes were utilized to yield the crude gold powder with high purity from the second leachate (adsorption, pre-elution of impurities, gold elution, and gold EW). Additionally, the remaining glutamate in the leachate and activated carbon could be reused for the next batch.

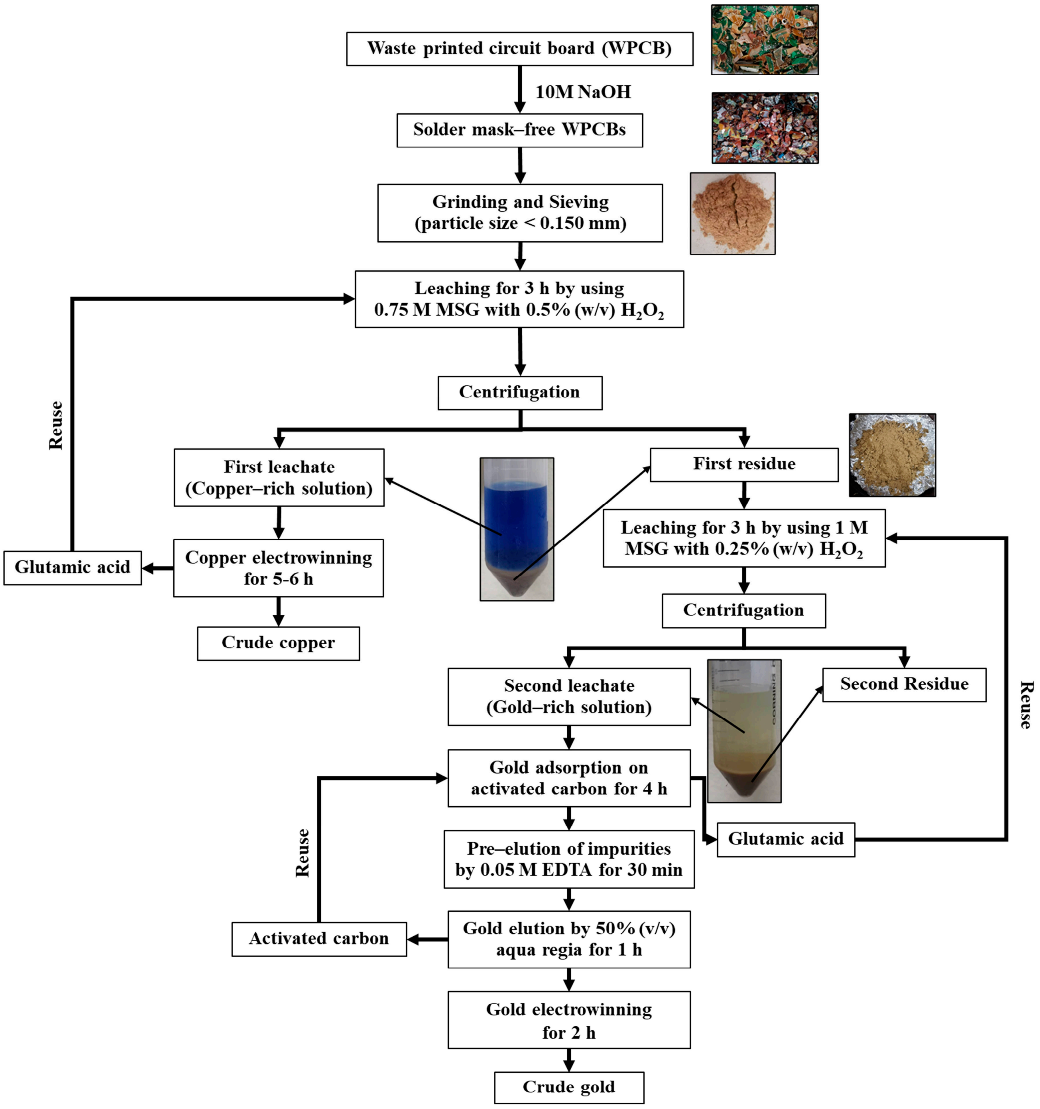

Figure 15. The overall process for recovery of copper and gold from waste printed circuit board.

This study proposed a complete process to recover copper and gold from WPCB using MSG supplemented with H_2O_2 as a lixiviant and relatively eco-friendly downstream process, as mentioned above. Although the WPCB used in this work appeared to be a low-grade WPCB with small amounts of precious metals and high base metal content [58], we believed that this process was of the potential to be further developed into the recovery of precious metals, namely gold from a mid-grade or high-grade WPCB, which requires further studies. Likewise, further study on the effect of centrifugation on the results may also be required as it may probably affect metal dissolution in particular. Toifl et al. (2003) reported that soil type was the most significant parameter in the total dissolved phosphorus extraction from the soil, while centrifugal speed and time played a marginal role [59]. Further, in soil moisture extraction, moisture recovery was influenced by a centrifugal force rather than time, whereas the solute concentration remained mostly constant with increasing recovery [60]. Imoto et al. (2018) found that centrifugal speed influenced the metal concentration during soil leaching tests [61]. The results obtained in this study provide a basis for future development of a larger scale and, eventually, commercial practices of metal recovery from E-waste.

The highly selective copper extraction and recovery was an essential advantage of this scheme, which promoted efficient utilization and industrial-level development to maximize economic and environmental benefits. Even though the leaching of gold was satisfactory, it was contaminated with other unwelcome metals. Our purification processes offered an acceptable outcome. Nevertheless, to enhance commercial prospects, establishing a perfectly capable decontamination approach remains a challenge. It could be probably improved by using other reported pre-elution methods, such as the Elutech process (using H_2SO_4 in the presence of H_2O_2) and chloride elution-based process [62]. In addition, further trials focusing on the reusability of chemicals or materials such as glutamate, activated carbon, and stripping agents are also needed.

4. Conclusions

The current research purposed to achieve the recovery of copper and gold from the WPCB using MSG-based lixiviant coupled with H_2O_2 as a clean oxidizer. The results indicated that the elimination of the solder mask layer was desired to increase the leaching efficacy. The maximum extractions of copper and gold were obtained by two stages of leaching under mild conditions (30 °C, 150 rpm, and initial pH of lixiviant of 7.00).

In the first leaching, copper was dissolved greatly (93%) with low contamination of other metals by using 0.75 M MSG coupled with 0.5% (w/v) H_2O_2.

In the second leaching, the gold extraction was maximized (86%) from the solid residue using 1 M MSG supplemented with 0.25% (w/v) H_2O_2.

Subsequently, 98–100% copper can be directly electrowon from the first leachate within 5–6 h. On the other hand, due to the contamination of other unfavorable metals in the second leachate, the gold recovery from this solution greatly required separation and purification before EW (100% gold electrodeposition within 2 h) as follows: the separation of gold from the second leachate by activated carbon (98% adsorption at 4 h), the pre-elution of contaminates by 0.05 M EDTA (no gold desorption), and the elution of gold from pre-eluted gold-loaded activated carbon using a diluted aqua-regia (99% gold stripping). This novel scheme could not only achieve the economically sustainable recovery of copper and gold without the reliance on traditional mining methods or dirty mining practices but also open doors to new opportunities to step forward into the next level of the cleaner hydrometallurgical process.

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