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Thiosulphate Leaching of Gold from Waste Mobile Phone Printed Circuit Boards: Influence of Key Chemical Agents on Leaching Behaviour

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Abstract

The Gold leaching from waste mobile phone printed circuit boards (PCBs) using an ammoniacal thiosulphate system was systematically investigated, with particular emphasis on the roles of dissolved oxygen, copper(II) ions, thiosulphate, and ammonia. Leaching experiments were conducted at 30 °C using dissolved oxygen and copper(II) ions acting as oxidants. The effects of reagent concentrations on gold dissolution kinetics were quantified using initial rate analysis. In the absence of Cu(II), the gold leaching rate exhibited an apparent reaction order of approximately 0.3 with respect to dissolved oxygen. Under oxygen-free conditions, reaction orders of 1.2, 0.38, and 0.32 were obtained for thiosulphate, Cu(II), and ammonia, respectively. A maximum gold dissolution of 98% within 10 min was achieved using a solution containing 60 mM thiosulphate, 10 mM Cu(II), and 0.26 M ammonia. Surface and phase characterisation by scanning electron microscopy (SEM) and X-ray diffraction (XRD) confirmed complete gold removal under optimal conditions. The results provide a coherent basis for optimising thiosulphate-based gold recovery from waste mobile phones.

Keywords: Electronic waste, gold, printed circuit boards, recycling, thiosulphate leaching.

1. Introduction

The electronics industry has experienced rapid growth over the past decades, accompanied by shortened product life cycles and accelerated technological obsolescence. As a consequence, waste electrical and electronic equipment (WEEE) has become one of the fastest-growing waste streams worldwide, with annual generation estimated at 20–50 million tonnes [1-3]. Improper management of this waste poses serious environmental and health risks, while at the same time representing a significant loss of valuable secondary resources. Gold is extensively used in electronic devices owing to its superior electrical conductivity, corrosion resistance, and chemical stability, particularly in printed circuit boards (PCBs) where it serves as a contact and connector material. Typical PCB scraps contain gold at concentrations far exceeding those of primary ores, for example approximately 31 ppm Au in personal computer boards [3-5]. Consequently, waste PCBs have been widely recognised as an attractive secondary source of gold, and gold recovery has been identified as the principal economic driver for PCB recycling [2, 4]. The development of efficient and environmentally benign gold recovery technologies is therefore of both economic and environmental importance.

Conventional gold extraction from electronic waste has largely relied on cyanide-based leaching processes. Although effective, cyanidation suffers from severe environmental and safety concerns, which have motivated extensive research into alternative lixiviants. Among these, thiosulphate leaching has emerged as one of the most promising non-cyanide technologies for gold recovery [3, 5-7]. In ammoniacal thiosulphate systems, gold dissolution is facilitated by copper(II) ions acting as redox catalysts, leading to the formation of stable gold(I) thiosulphate complexes, as shown in (1). However, the overall leaching behaviour is governed by a complex network of coupled reactions, including thiosulphate oxidation (2), copper redox cycling, ammonia complexation, and the involvement of dissolved oxygen (3) [3, 5–7].

$$Au + Cu(NH_3)_4^{2+} + 5S_2O_3^{2-} \rightarrow Au(S_2O_3)_2^{3-} + Cu(S_2O_3)_3^{5-} + 4NH_3 \quad (1)$$

$$2Cu(NH_3)_4^{2+} + 8S_2O_3^{2-} \rightarrow 2Cu(S_2O_3)_3^{5-} + S_4O_6^{2-} + 8NH_3 \quad (2)$$

$$2\,Cu(S_2O_3)_3^{5-} + 8NH_3 + 1/2O_2 + H_2O \rightarrow 2Cu(NH_3)_4^{2+} + 2OH^- + 6S_2O_3^{2-} \quad (3)$$

Numerous studies have investigated gold leaching in ammoniacal thiosulphate media, focusing on aspects such as reaction mechanisms, kinetics, and the effects of individual parameters including oxygen, copper(II) ions, thiosulphate, ammonia, and temperature [6-9].

Gold leaching from waste mobile phone PCBs using this system has also been reported [10, 11, 13]. V.H. Ha *et al.* [10] achieved 90% gold recovery within 10 hours using a solution containing 20 mM copper, 0.12 M thiosulphate, and 0.2 M ammonia. Similarly, A. Tripathi *et al.* [11] reported leaching efficiencies of 56.7% and 78% for shredded and intact PCBs, respectively, under optimized conditions of 0.1 M ammonium thiosulfate,

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40 mM $CuSO_4$, pH 10–10.5, 10 g/L pulp density, and 250 rpm stirring speed at room temperature over 8 hours.

While these studies have provided valuable insights, most investigations have examined the influence of single variables under fixed conditions. As a result, the reported data are often fragmented, and a systematic understanding of how key leaching parameters interact with one another remains limited. In particular, the combined and sometimes competing roles of dissolved oxygen, copper(II) concentration, and thiosulphate stability have not been comprehensively quantified for waste mobile phone PCBs, which are among the richest secondary sources of gold. The lack of integrated kinetic analysis hampers the identification of optimal operating windows that balance high gold dissolution rates with acceptable thiosulphate consumption and copper stability. Therefore, the objective of the present study is to systematically evaluate the individual and interactive effects of dissolved oxygen, thiosulphate concentration, copper(II) ions, and ammonia on the leaching kinetics and efficiency of gold from waste mobile phone PCBs. By employing initial rate analysis and controlled experimental conditions, this work aims to clarify the roles of key chemical agents in ammoniacal thiosulphate leaching and to provide a more coherent basis for process optimisation in the recycling of gold from electronic waste.

2. Experiments

2.1. Materials and Reagents

Waste mobile phone PCBs were obtained from a PCB manufacturer in Korea. The average gold content was approximately 440 g tonne^{-1}, with an exposed gold contact area of 486 mm² per PCB plate. Cross-sectional SEM–EDS analysis revealed a multilayer structure consisting of a gold–nickel surface layer over nickel, copper, and polymer substrate layers. The outermost layer contained more than 90 wt.% gold, indicating minimal interference from other metals such as nickel and copper during initial leaching (Fig. 1).

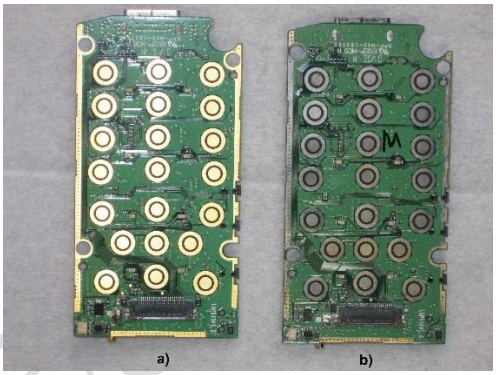
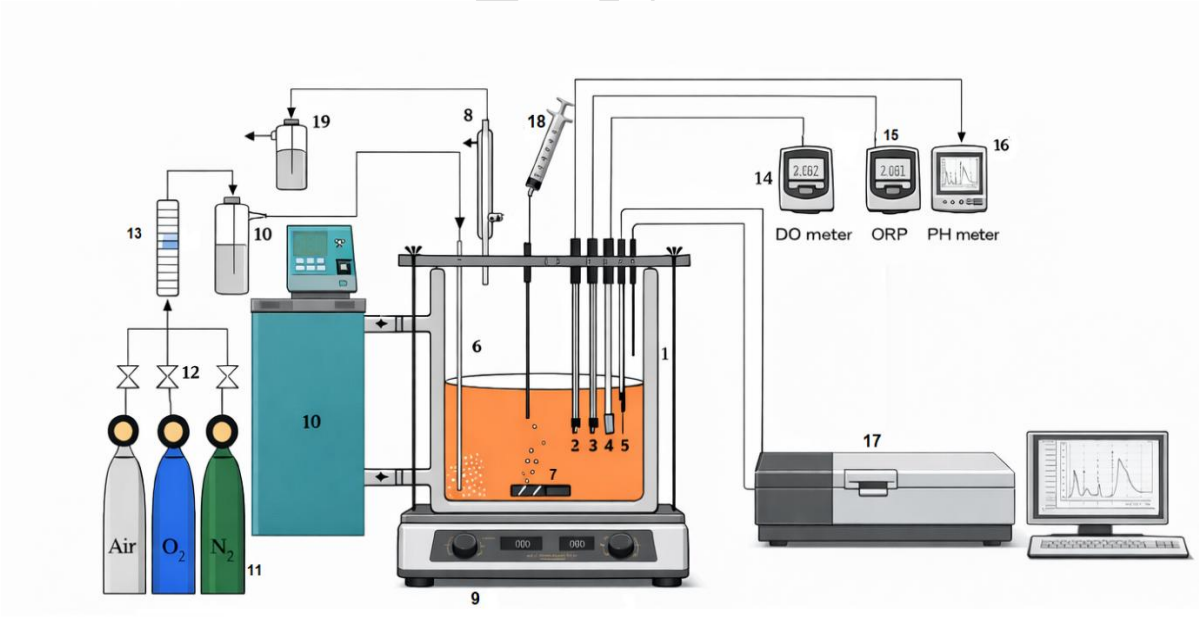


Fig. 1. PCB samples a) before leaching and b) after leaching



1. Reactor
2. pH electrode
3. ORP electrode
4. DO electrode
5. Inlet tube of UV/vis Spectrophotometer
6. Gas sparger
7. Magnetic stirrer bar
8. Condenser
9. Magnetic stirrer
10. Thermostatic water bath
11. Gas cylinders
12. Valves
13. Flow meter
14. DO meter
15. ORP meter
16. pH meter
17. UV/vis Spectrophotometer
18. Cylinder for sampling
19. Gas scrubber

Fig. 2. A schematic diagram of reactor system for gold leaching

Analytical-grade sodium thiosulphate, ammonium sulphate, sodium chloride, aqueous ammonia (25 wt.%), and copper sulphate were used. All solutions were prepared with deionised water. Unless otherwise stated, the molar ratio of ammonia to ammonium ions was maintained close to unity, and the solution pH was approximately 10 (measured at 20 °C).

2.2. Leaching Experiments

The leaching experiments were conducted using intact PCBs to minimize the influence of nickel and copper compared to shredded PCBs [11]. Therefore, intact PCB samples were selected for this study. Leaching tests were performed in a 0.5 L jacketed glass reactor equipped with magnetic stirring at 400 rpm (Fig. 2). Each experiment used 400 mL of leaching solution and ten PCB plates, which were suspended using nylon threads. The temperature was maintained at 30 ºC within ±0.5 °C. Dissolved oxygen concentrations were controlled by bubbling nitrogen, air, or oxygen through the solution. To minimise ammonia loss, inlet gases were pre-saturated with ammonia. Gold dissolution was monitored by periodic sampling, and initial leaching rates were determined based on gold dissolution per unit surface area under conditions of negligible reagent depletion.

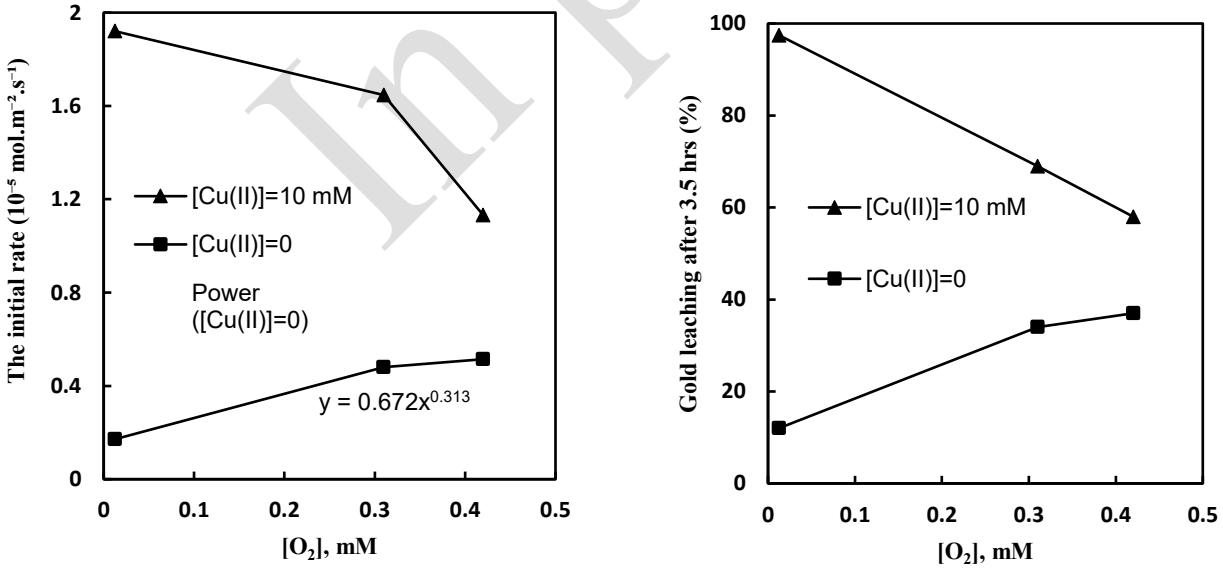
2.3. Analytical Methods

Gold concentrations in solution were determined by atomic absorption spectroscopy (AAS), while residual gold in solids was analysed following aqua regia digestion. Thiosulphate concentrations were measured by iodometric titration, with acetic acid added to suppress copper–ammonia interference. Cu(II) concentrations were monitored by Ultraviolet-visible (UV–Vis) spectrophotometry at 608 nm. pH, redox potential, temperature, and dissolved oxygen were continuously recorded. Scanning Electron Microscopy paired with Energy-Dispersive X-ray Spectroscopy (SEM–EDS) was used for surface characterisation before and after leaching.

3. Results and Discussion

3.1. Effect of Dissolved Oxygen Concentration

The results presented in Fig. 3 demonstrate that dissolved oxygen (DO) concentration has a significant influence on both the initial leaching kinetics and the overall gold leaching performance, with a clear distinction between systems with and without the presence of Cu(II). In the absence of Cu(II), oxygen acted as the sole oxidant and increasing DO enhanced the initial gold leaching rate from 0.17 to 0.51 × 10^{-5} mol m^{-2} s^{-1} (Fig. 3a). The experimental relationship between the reaction rate and oxygen concentration follows a power-law expression ($r = 0.672[O_2]^{0.313}$), indicating that the leaching process is partially governed by diffusion and oxygen availability in the solution. Accordingly, higher DO levels result in an improved gold extraction after 3.5 h of leaching (Fig. 3b).



(a) (b)

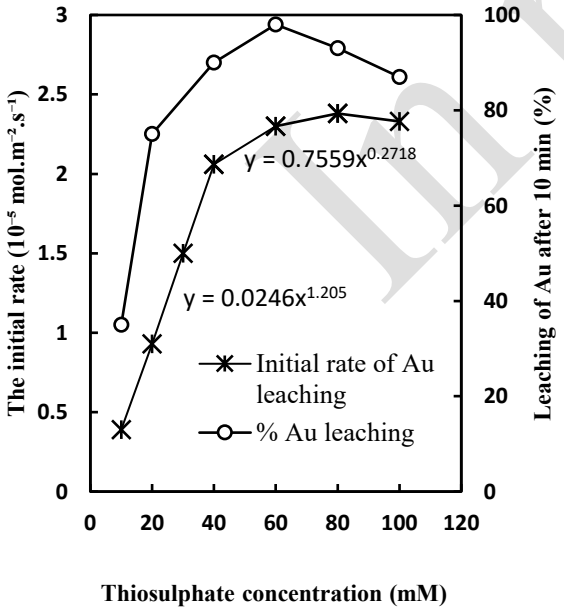
Fig. 3. Effect of disolved oxygen concentraition on gold leaching: (a) initial leaching rate and (b) leaching performance. Experimental conditions: 60 mM $(NH_4)_2S_2O_3$; 0.26 M NH_3; 30 °C; 10 mL/min flow rate by nitrogen bubbling (DO ≈ 0.012 mM), air bubbling (DO ≈ 0.3 mM) and oxygen bubbling (DO ≈ 0.42 mM)

In contrast, in the presence of 10 mM Cu(II), dissolved oxygen has a negative effect on both the initial gold leaching rate and the overall gold extraction efficiency. As the DO concentration increased from 0.012 to 0.42 mM, the gold leaching rate decreased from 1.92×10^{-5} to 1.13×10^{-5} mol m^{-2} s^{-1}. This behaviour can be attributed to the simultaneous presence of oxygen and Cu(II), which leads to thiosulphate depletion in solution [8] and a reduction in the Cu(II)/Cu(I) concentration ratio, thereby decreasing the redox potential of the leaching solution [9]. Overall, these results demonstrate that the optimal dissolved oxygen concentration strongly depends on the presence of Cu(II). While oxygen enhances gold leaching in the absence of Cu(II), lower dissolved oxygen levels are more favorable for achieving improved leaching kinetics and efficiency in Cu(II)-assisted gold leaching.

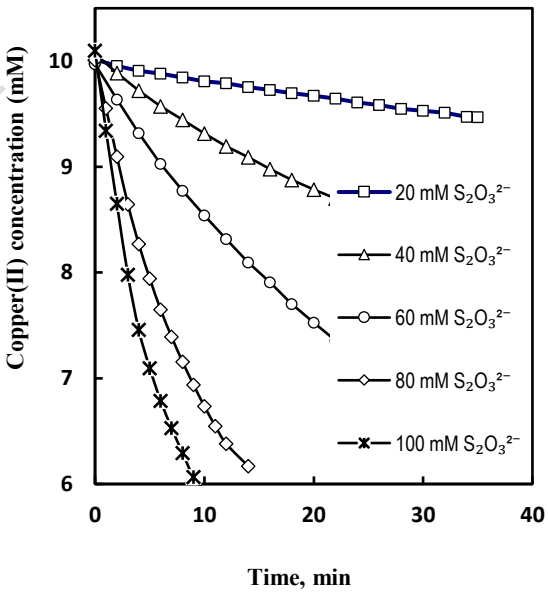
3.2. Effect of Thiosulphate Concentration

In ammoniacal cupric–thiosulphate systems, gold dissolution proceeds through coupled oxidation and complexation reactions, with thiosulphate stabilising Au(I) as the $Au(S_2O_3)_2^{3-}$ complex (1). According to the copper-catalysed mechanism, thiosulphate participates in both anodic gold oxidation and cathodic $Cu(NH_3)_4^{2+}$ reduction, while being simultaneously consumed through copper-catalysed oxidation reactions [9]. Fig. 4(a) illustrates the effect of thiosulphate concentration (10–100 mM) on the initial leaching rate and the percentage of gold dissolution after 10 min under oxygen-free conditions, while the corresponding variation of Cu(II) concentration is shown in Fig. 4b.

As shown in Fig. 4a, the initial gold leaching rate increases with increasing thiosulphate concentration at lower concentrations, indicating enhanced formation and stabilization of the $Au(S_2O_3)_2^{3-}$ complex. However, further increases in thiosulphate concentration result in a gradual decline in the leaching rate. The reaction order with respect to thiosulphate concentration was determined to be 1.205 in the range of 10–40 mM and decreased to 0.2718 in the range of 40–100 mM, suggesting a transition in the rate-controlling mechanism. The maximum gold dissolution of approximately 98% after 10 min was achieved at a thiosulphate concentration of 60 mM. Although higher thiosulphate concentrations favor the stability of the $Au(S_2O_3)_2^{3-}$ complex, the Cu(II) concentration profiles in Fig. 4b reveal that increasing thiosulphate concentration accelerates the reduction of Cu(II), as shown in (2), leading to a lower steady-state Cu(II) concentration and a decreased Cu(II)/Cu(I) concentration ratio. This shift in the redox balance diminishes the oxidizing capacity of the leaching system, thereby inhibiting gold dissolution, as discussed in Section 3.1.

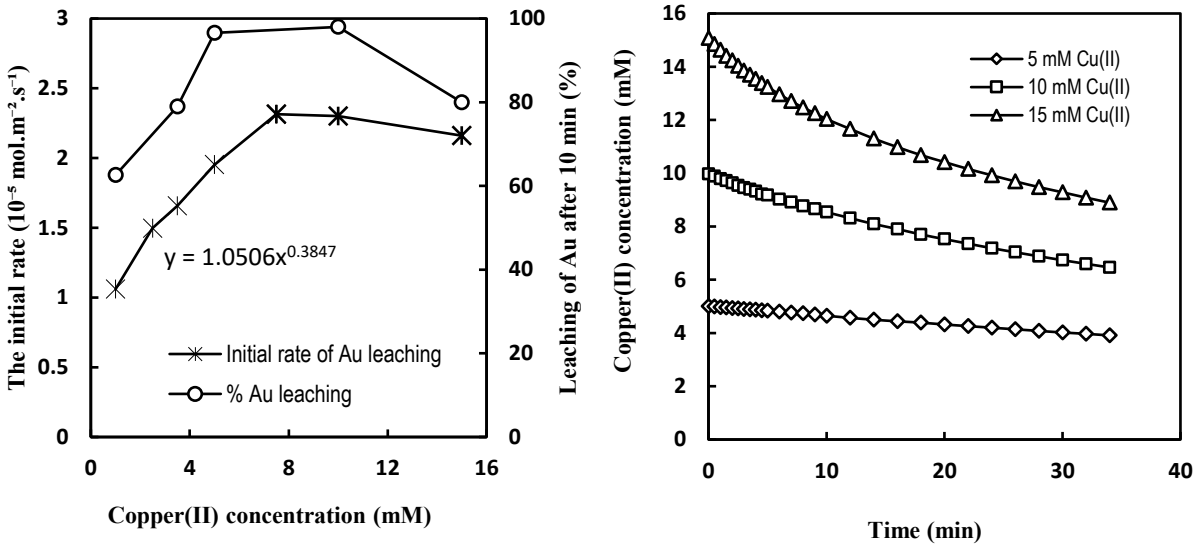


(a)



(b)

Fig. 4. Effect of thiosulphate concentration on (a) the initial rate and performance of gold leaching and (b) the remainder of copper(II) concentration in leach solution. Experimental condititons: 10 - 100 mM $Na_2S_2O_3$; 10 mM CuS_2O_4; 0.26M NH_4Cl; 0.26 M NH_3; nitrogen sparged at 10 mL/min; 30 °C; 400 rpm



(a) (b)

Fig. 5. Effect of copper(II) ions concentration on (a) the initial rate and performance of gold leaching and (b) the copper(II) ion concentration. Experimental condititons: 1 - 15 mM CuS_2O_4; 60 mM $Na_2S_2O_3$; 0.26M NH_4Cl; 0.26 M NH_3; 10 mL/min flow rate by nitrogen bubbling; 30 °C; 400 rpm

3.3. Effect of Copper(II) ion Concentration

The role of Cu(II) ions in the oxidation of metallic gold to Au(I) species is described in (1). In ammoniacal thiosulphate systems, the presence of Cu(II) significantly enhances the rate of gold dissolution, although excessive copper concentrations are known to accelerate thiosulphate consumption through catalytic oxidation, as shown in (2) [7]. Therefore, an optimum Cu(II) concentration is required to maximize gold leaching while minimizing reagent degradation.

Fig. 5a illustrates the effect of Cu(II) concentration (1–15 mM) on the initial leaching rate and gold extraction after 10 min under otherwise identical conditions. At low Cu(II) concentrations (1–7.5 mM), increasing Cu(II) markedly enhances the gold leaching rate, with an apparent reaction order of approximately 0.3847 with respect to Cu(II). This trend indicates that gold dissolution is promoted by increased availability of cupric species participating in the redox cycle. However, at higher Cu(II) concentrations (>7.5 mM), further increases in Cu(II) result in a gradual decrease in the leaching rate. A similar trend is observed for gold extraction after 10 min, which increases from 62.6% to 96.6% as the Cu(II) concentration rises from 1 to 5 mM, remains nearly constant in the range of 5–10 mM, and then decreases to 80.3% at 15 mM Cu(II). These results indicate that effective gold leaching is achieved at Cu(II) concentrations in the range of approximately 7.5–10 mM. According to the copper-catalyzed mechanism for gold leaching in ammoniacal thiosulphate systems, $Cu(NH_3)_4^{2+}$ species act as electron acceptors at the cathodic sites on the gold surface [8]. At low Cu(II) concentrations, the leaching rate is limited by the diffusion of cupric species to the gold surface, and increasing Cu(II) therefore enhances both the oxidation rate and leaching efficiency of gold. Once the Cu(II) concentration becomes sufficiently high that its mass transport is no longer rate-limiting, this positive effect diminishes.

In addition, Fig. 5b shows the temporal evolution of Cu(II) concentration under the same conditions. The results indicate that the rate of Cu(II) reduction increases strongly with increasing initial Cu(II) concentration and follows an apparent second-order dependence. At Cu(II) concentrations above 10 mM, the rapid reduction of cupric ions leads to accelerated consumption of both Cu(II) and thiosulphate, which are essential for sustained gold leaching. Consequently, excessive Cu(II) concentrations reduce both the leaching rate and overall gold extraction efficiency.

3.4. Effect of Ammonia Concentration

The effect of ammonia concentration (0.15–0.30 M) on gold leaching is shown in Fig. 6. Increasing the ammonia concentration from 0.15 to 0.26 M enhanced the initial gold leaching rate from 1.65 to 1.95×10^{-5} mol m^{-2} s^{-1}, corresponding to a reaction order of 0.318 with respect to ammonia. The maximum leaching rate was observed at 0.26 M NH_3, while further increases in ammonia concentration resulted in a gradual decline in the leaching rate (Fig. 6a).

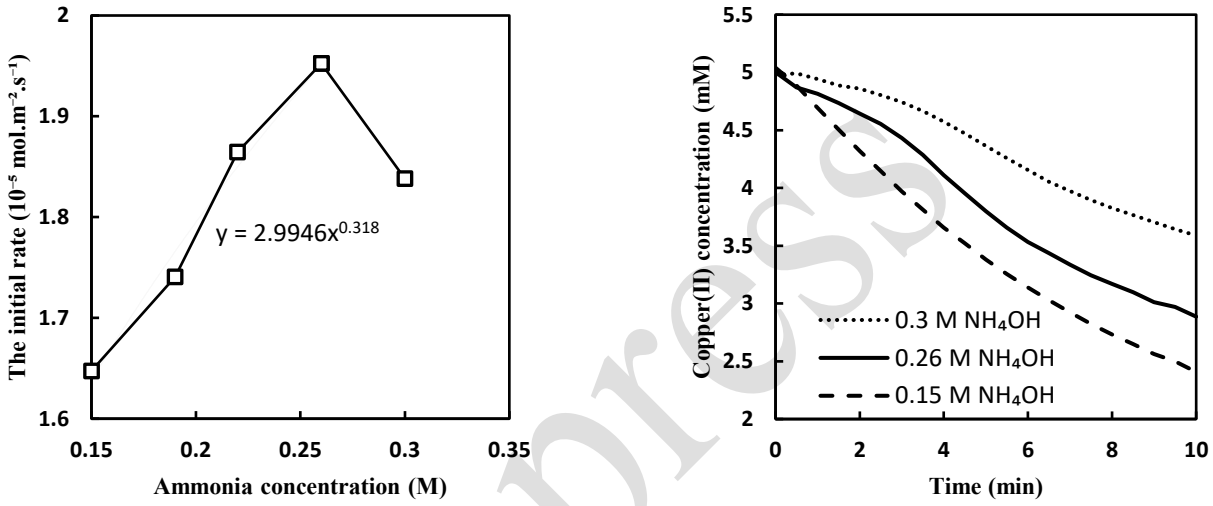
Ammonia plays a critical role in stabilising Cu(II) as the cupric tetra-ammine complex, $Cu(NH_3)_4^{2+}$, which acts as the primary oxidant for gold dissolution in thiosulphate solutions. This effect is supported by UV–visible spectrophotometric measurements

(Fig. 6b), which show higher residual concentrations of Cu(II) complexes at elevated ammonia levels. In addition, ammonia suppresses gold passivation by forming the aurous diammine complex, $Au(NH_3)_2^+$, which subsequently reacts with thiosulphate to produce the more stable $Au(S_2O_3)_2^{3-}$ complex [12]. At higher ammonia concentrations, however, the equilibrium of the reaction shifts towards $Au(NH_3)_2^+$, reducing the availability of $Au(S_2O_3)_2^{3-}$ and thereby decreasing the gold leaching rate.

The optimum ammonia concentration is therefore system-dependent, with 0.26 M NH_3 being optimal for solutions containing 60 mM thiosulphate and 10 mM Cu(II). Under these conditions, the process achieved an initial gold leaching rate of 2.3×10^{-5} mol m^{-2} s^{-1} and 98% dissolution within 10 min, significantly outperforming the leaching rate reported by O. Sitando *et. al.* (1.7×10^{-5} mol m^{-2} s^{-1} [13]) and the dissolution efficiency reported by A. Tripathi *et al.* (78% within 8 h [11]).

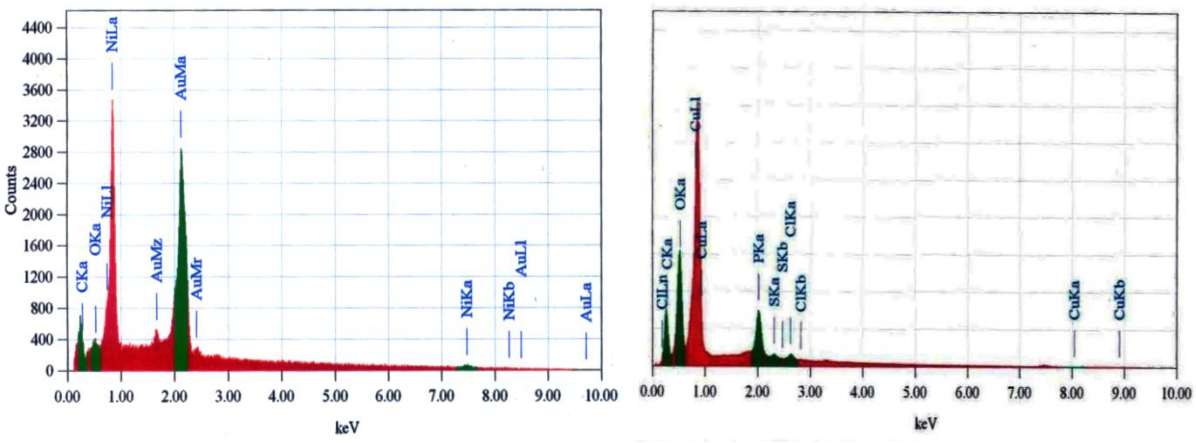
3.5. Surface Characterisation

SEM–EDS analysis confirmed complete removal of gold from the PCB surface under optimal leaching conditions, with no detectable gold remaining in the residue (Fig. 7).



(a) (b)

Fig. 6. Effect of ammonia concentration on (a) the initial rate of gold leaching and (b) copper(II) reduction. Experimental condititons: 0.15 - 0.3 M NH_3; 5 mM CuS_2O_4; 60 mM $Na_2S_2O_3$; 10 mL/min flow rate by nitrogen bubbling; 30 °C; 400 rpm



(a) (b)

Fig. 7. Energy dispersive spectrum of samples a) before leaching and b) after leaching

4. Conclusions

The results presented in this study highlight the complexity of the chemistry in the ammonia–thiosulphate–copper system, particularly in the presence of oxygen, and its critical role in gold leaching from PCBs. In the absence of Cu(II) ions, the gold leaching rate was found to be relatively low. In contrast, when Cu(II) ions were present, dissolved oxygen accelerated thiosulphate oxidation and Cu(II) reduction, thereby decreasing the concentrations of both species, which are essential for sustained gold leaching. Thiosulphate, ammonia, and Cu(II) ions exhibited a positive influence on the gold leaching rate at low concentrations, but a negative effect at higher concentrations. The inhibitory effects of high thiosulphate and Cu(II) concentrations, together with the beneficial effect of lower ammonia concentration, are likely associated with changes in the Cu(II)/Cu(I) concentration ratio and the resulting redox conditions of the leaching system. Therefore, achieving a sustainable and efficient gold leaching process while maintaining acceptable thiosulphate consumption requires a careful balance of operating conditions. Based on the results of this study, the optimal conditions were identified as 0.26 M ammonia, 60 mM thiosulphate, and 10 mM Cu(II).

Acknowledgments

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