The recovery of gold from copper-rich Zimbabwean gold ore by cold stripping

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Gold ore grade has declined internationally and the mineralogy is now very complex. Therefore, the utilisation of low-grade ores that include ore with high copper levels is now a necessity. Copper cyanide complexes that are competitively adsorbed with gold onto activated carbon are formed during the cyanidation process of such ores. A series of copper adsorption experiments were conducted to decrease the amount of copper cyanide complexes reporting to the pregnant electrowinning electrolyte. Tests on selective copper cold stripping were also conducted to selectively elute copper from loaded head carbon before the elution process. The results showed that increasing the solution pH from 9 to 11 decreased the copper adsorption from about 15.1% to 8.2%, but in contrast, the gold adsorption decreased from 98.7% to 95.1%. The optimal determined cold stripping parameters were 2% cyanide strength, 1% sodium hydroxide, 27 g/L carbon concentration, and four hours residence time. This experimental procedure effectively reduced the amount of copper in the gold stripping process and has the potential to be used in large-scale applications.

Keywords: Gold cyanide, copper cyanide, cold stripping, adsorption, desorption, and elution.

INTRODUCTION

According to Medina and Anderson, the gold ore grade worldwide has been declining and has grown increasingly complicated and diversified. Because of this, the only option available to gold miners and processors is to investigate ways to use low-grade ores. Since cyanidation of gold ores poses several challenges, it is necessary to examine solutions for related issues (Medina & Anderson, 2020). A 1.53 g/t gold ore that is high in copper is processed on a certain gold mine in Zimbabwe. Processes such as crushing, milling, dewatering, leaching, elution, electrowinning, and smelting are used to process run-of-mine ore as shown in Figure 1.

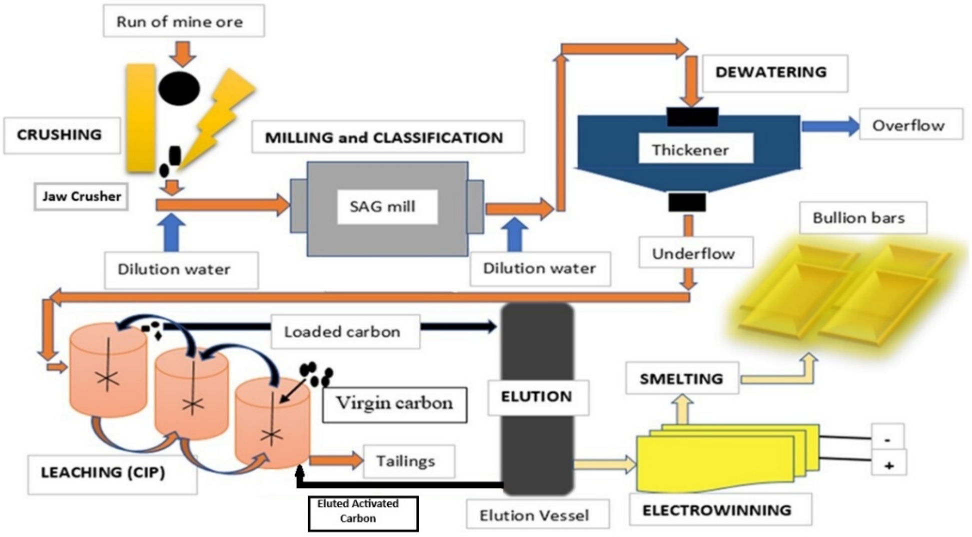

Figure 1. Typical gold processing flow sheet (Muchada H, et al.*, 2024)***.**

Copper combines with cyanide during the intense cyanidation of gold to produce copper cyanide complexes, which include Cu $(CN)_2^-$, Cu $(CN)_3^{2-}$, and Cu $(CN)_4^{3-}$. In a low pH environment, the copper dissolving effect is amplified, leading to an increase in cyanide consumption. (Adams, 2005) Since copper is the main component of run-of-mine ore, its influence on the leaching process should be reduced. Figure 2 shows the head assays of gold ore from different underground mining sections. These gold ore samples contain high amounts of copper hence the classification copper-rich Zimbabwean gold ore.

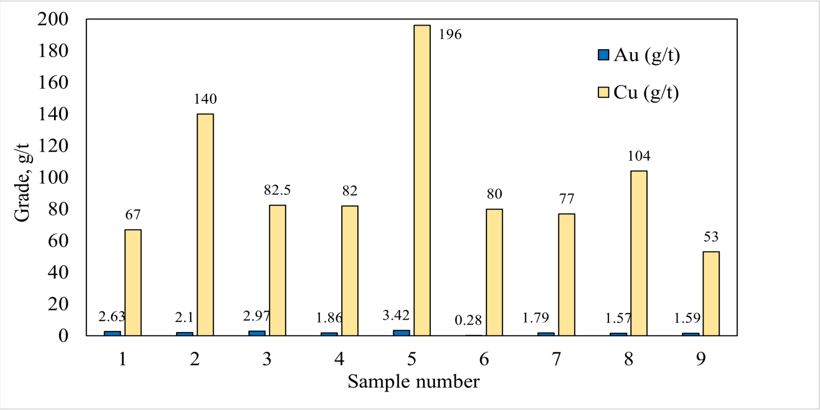

Figure 2. Head assays of gold ore from different underground mining sections near the processing plant (Muchada H, et al.*, 2024)***.**

Silver (Ag), copper (Ag), arsenic (As), antimony (Sb), bismuth (Bi), iron (Fe), lead (Pb), zinc (Zn), and bismuth (Bi) are among the metals that are commonly associated with gold, (Grover & Okewale, 2023). Thus, four groups of minerals that contain gold can be identified as copper sulfides, silver-rich ores, arsenic sulfides, and antimony sulfides.

Copper is commonly detected in certain ores as bornite, chalcocite, tennanitte, tetrahedrite, and chalcopyrite, accounting for around 20% of all gold ore deposits. Gold typically coupled with copper sulfide alters the process whereby gold is extracted. Chalcopyrite ($CuFeS_2$), the most common copper mineral, has a copper content of approximately 34.5%. It can undergo oxidation to generate chalcocite (Cu_2S), hematite (Fe_2O_3), or covellite (CuS). Copper content in chalcocite is 79.8% and in vellite, 66.4%, respectively (Deschenes *et al.*, 2012). These substances are found in secondary enrichment zones and are the consequence of primary copper sulfide alteration. Bornite (Cu_5FeS_4) is another major copper sulfide ore (Marsden & House, 2006).

According to Equations 1– 3, copper minerals were observed to dissolve in 0.1% sodium cyanide (Gupta & Mukherjee, 1990).

$$\mathrm{2CuO + 7NaCN + H_2O \leftrightarrow 2Na_2Cu(CN)_3 + 2NaOH + NaCNO} \quad [1]$$

$$\mathrm{Cu_2S + 6NaCN \rightarrow 2Na_2Cu(CN)_3 + Na_2S} \quad [2]$$

$$\mathrm{CuFeS_2 + 19NaCN + 2NaOH \rightarrow 2Na_2Cu(CN)_3 + 4Na_2S + 2Na_4Fe(CN)_6 + NaCNO + H_2O} \quad [3]$$

With the exception of chalcopyrite, the majority of copper-containing minerals are highly cyanide soluble. Copper is problematic because, during the cyanidation process of copper-rich gold ores, the cyanide that is expected to react with gold is also consumed by copper (Equations 1–3), resulting in low gold extraction. Other issues with copper include competing adsorption and decreased electrowinning cell efficiency. Gold can also be lost due to the cementation of copper minerals (Vegter & Sandenbergh, 1992).

Copper Cyanide Chemistry

The copper complexes are in many forms in a copper cyanide system, and the cyanidation reactions can be defined as follows. Equations 4–7 (Vogel, 1978).

$$\mathrm{Cu^+ + CN^- \rightarrow CuCN}(s) \quad [4]$$

$$\mathrm{CuCN}(s) + \mathrm{CN^- \rightarrow Cu(CN)_2^-} \quad [5]$$

$$\mathrm{Cu(CN)_2^- + CN^- \rightarrow Cu(CN)_3^{2-}} \quad [6]$$

$$\mathrm{Cu(CN)_3^{2-} + CN^- \rightarrow Cu(CN)_4^{3-}} \quad [7]$$

$$\mathrm{Cu(CN)_4^{3-} + 2H^+ \rightarrow Cu(CN)_2^- + 2HCN} \quad [8]$$

$$\mathrm{Cu(CN)_3^{2-} + H^+ \rightarrow Cu(CN)_2^- + HCN} \quad [9]$$

The Eh-pH diagram of the copper-cyanide-water system in Figure 3 demonstrates that the Cu $(CN)_3^{2-}$ complex dominates the cyanidation environment. In the presence of free cyanide, the copper complex will react with cyanide according to Equation 2. Copper cyanide (Cu $(CN)_3^{2-}$) dominates in alkaline pH environments with somewhat high free cyanide levels (Vogel, 1978). Cu $(CN)_2{}^-$ complex formation is strongly preferred in a low pH, free cyanide environment. This occurrence can be described by the reaction shown in Equations 8 and 9:

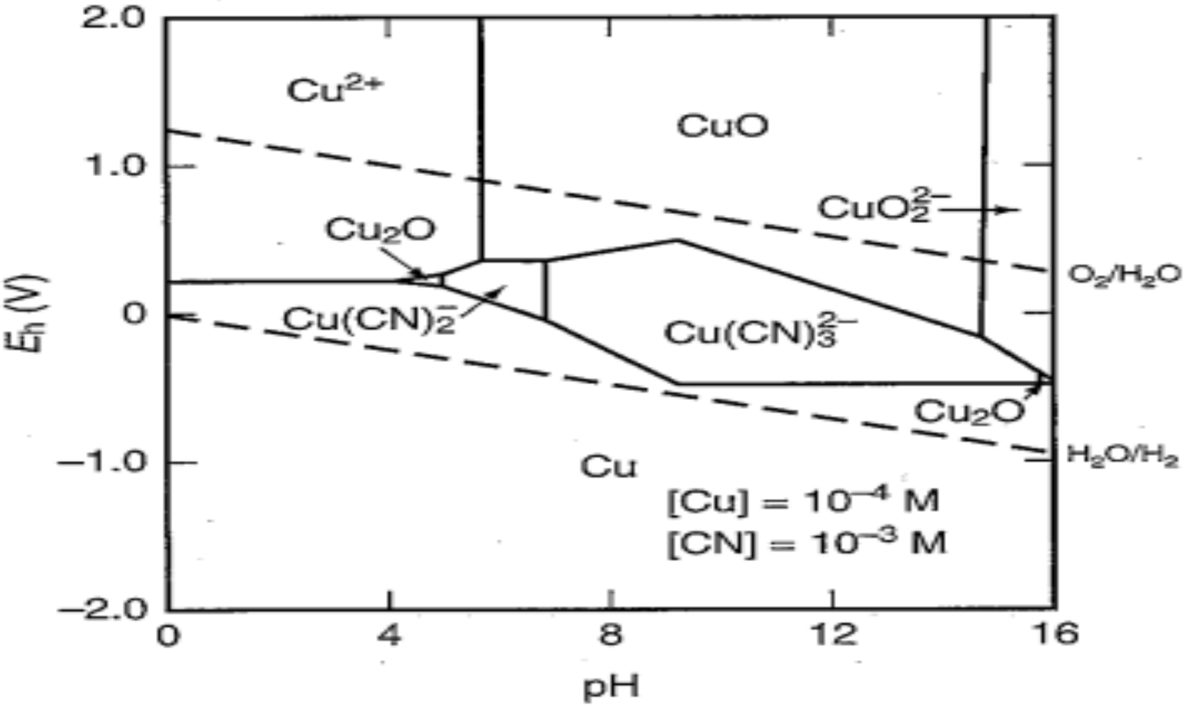

Figure 3. Eh-pH diagram for copper-cyanide-water system at 25°C (Simons, et al.*, n.d.)*·

Copper was also shown to dissolve in cyanide during the leaching of copper-rich gold ores, forming soluble copper-cyanide complexes Cu $(CN)_2{}^-$, Cu $(CN)_3^{2-}$, and Cu $(CN)_4^{3-}$. The concentration and presence of these complexes in solution are determined by the concentration of free cyanide and pH (Oraby & Eksteen, 2015). In an environment with low free cyanide and pH (Nicol *et al.*, 1987), the Cu $(CN)_2{}^-$ and Cu $(CN)_3^{2-}$ complexes are prevalent and favoured. When the pH and free cyanide concentrations rise, the presence of copper as the Cu $(CN)_4^{3-}$ complex increases while copper adsorption on activated carbon decreases (Marsden & House, 2006).

The adsorption strength of the three copper complexes increases in the order:

$$\mathrm{Cu(CN)_4^{3-}} < \mathrm{Cu(CN)_3^{2-}} < \mathrm{Cu(CN)_2^{-}}$$

According to Marsden and House, keeping a 4:1 cyanide-to-copper molar ratio can limit copper adsorption on activated carbon. Alternatively, selective desorption of cold-stripped copper cyanide has been reported. As a result, pH and free cyanide levels must be closely monitored during a gold extraction operation from an ore high in cyanide-soluble copper (Marsden & House, 2006). Oladele (2015) indicated that if copper is present in the gold ore, flotation might be used to separate copper from gold. If flotation is not a possibility, gold can be obtained as a byproduct of copper smelting. If the copper values are inadequate to justify smelting, the copper-rich gold ore is cyanided (Oladele, 2015).

EXPERIMENTAL

The experiments were primarily concerned with limiting the amount of adsorbed copper cyanide and selectively eluting copper from loaded head carbon collected from header tanks in the carbon-in-leach section of Zimbabwean gold ore, to reduce the amount of copper in the electrowinning process.

Sample Collection and Preparation

Using a sample cutter, about 3 litres of pulp from a Zimbabwean gold ore graded at 1.53 g/t was retrieved from an actual plant leach tank adjacent to adsorption tanks and deposited in a 20-litre bucket. The pulp was then transferred to a steel pressure filter vessel and filtered. Three 3 mL head samples were obtained from the filtered solution and taken to the assay facility for gold and copper solution analysis using an atomic adsorption spectrometer. Another set of three 300 mL solutions was extracted from the filtrate and adjusted to pH levels of 9, 10.5, and 11 using sodium hydroxide and hydrochloric acid.

Copper Adsorption Test Procedure

Using the flow sheet shown in Figure 4, 4g of virgin activated carbon was added to a 300 ml loaded solution containing 1.29 mg/L Au and 16.26 mg/L Cu before placing the beaker on a magnetic stirrer. The magnetic stirrer and stopwatch were turned on simultaneously, and 3 mL of sample solution was drawn every 15 minutes for a total residence duration of one hour. After the experiment was finished, the samples were taken to the assay laboratory and examined for gold and copper with an atomic adsorption spectrometer. The process was repeated at least three times with different pH solutions made in sample preparation stage.

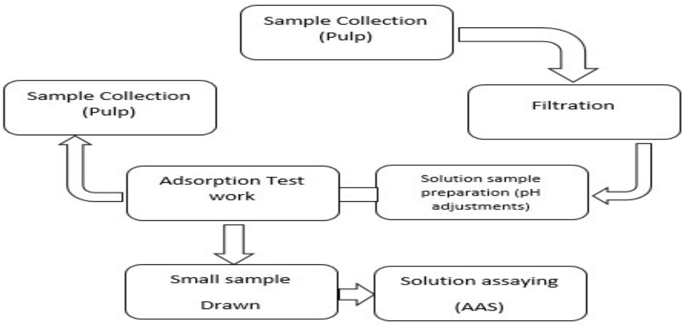

Figure 4. Flowsheet for copper adsorption experimental procedure (Muchada H, et al., *2024)* Copper Cold Stripping.

The copper and gold loaded activated carbon samples from another industrial head tank were collected and washed, placed on an 850-micron screen, and baked in an oven for approximately 45 minutes. After drying, the activated carbon was stored in clearly labeled sample holders. Then sodium hydroxide-cyanide solutions were prepared to obtain various concentrations, as shown in Table I.

Table I. Sodium hydroxide and cyanide concentration (Muchada H, et al., *2024)*

Sample Number	**Cyanide Strength (CN^-), %**	**Sodium Hydroxide Strength (OH^-), %**
1	0.1	1.0
2	0.4	1.0
3	0.8	1.0
4	1.5	1.0
5	2.0	1.0
6	2.5	1.0
7	3.0	1.0

Three samples of loaded activated carbon were taken to the assay laboratory for initial copper and gold analysis. The produced sodium hydroxide and cyanide solutions were poured into a 300-ml beaker, filled to the mark, and placed on a magnetic stirrer. Then 4 g of loaded activated carbon was added to the beaker containing the sodium hydroxide and cyanide solutions as given in Table I. Soon after the loaded activated carbon was added to the solution, the magnetic stirrer and timer were started simultaneously. After around two hours of residence time in the beaker, solution samples were extracted using a syringe. After the experiment, the stripped activated carbon was collected and analysed for copper and gold at the assay laboratory. The technique was repeated with varying sodium hydroxide strength according to Table II while maintaining the same residence time, cyanide strength, carbon mass, and solution volume.

Table II. Sodium hydroxide strength used in the investigation (Muchada H, et al., 2024)

Sodium hydroxide sample ID	Sodium hydroxide strength (%)
S1	1
S2	2
S3	3

To investigate the effects of loaded activated carbon concentration, the same method was repeated with the loaded activated carbon adjusted according to Table III while the sodium hydroxide, cyanide strength, residence time, and solution volume remained constant.

Table III. The loaded activated carbon masses used (Muchada H, et al.*, 2024)*

Carbon sample ID	Carbon mass (g)
C1	13
C2	20
C3	27
C4	33

To investigate the effects of resident time, the aforesaid procedure was used while varying the residence period by up to six hours while keeping the remaining parameters constant.

RESULTS AND DISCUSSION

Effect of pH on Copper Adsorption

Copper in solution was measured at various time intervals, and the pH of the solution was determined, as shown in Figure 5.

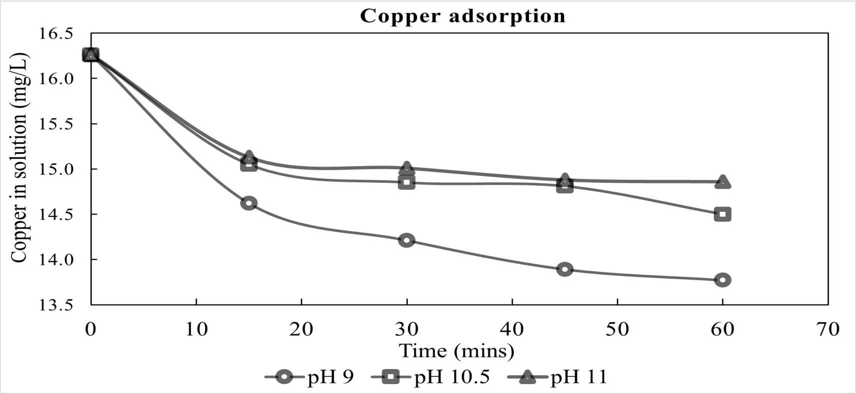

Figure 5. The relationship between the amount of copper in solution and time using virgin carbon (initial copper assay=16.62 mg/L, virgin carbon=4g and solution volume=300 ml) (Muchada H, et al.*, 2024).*

Generally, the amount of copper in the solution decreased over time, indicating that activated virgin carbon absorbed copper from the solution. However, the degree of adsorption varied depending on the pH of the solution.

Figure 5 shows that at pH 9, the initial high copper concentration gradient resulted in a fast adsorption rate of 5.47×10^{-7} g/s during the first 15 minutes of the experiment. After 45 minutes, the adsorption rate reduced to 9.4×10^{-8} g/s as the copper concentration gradient decreased over time. The copper in the solution was adsorbed and lowered to a final value of around 13.77 mg/L, achieving approximately 15.3% copper adsorption as shown in Figure 6.

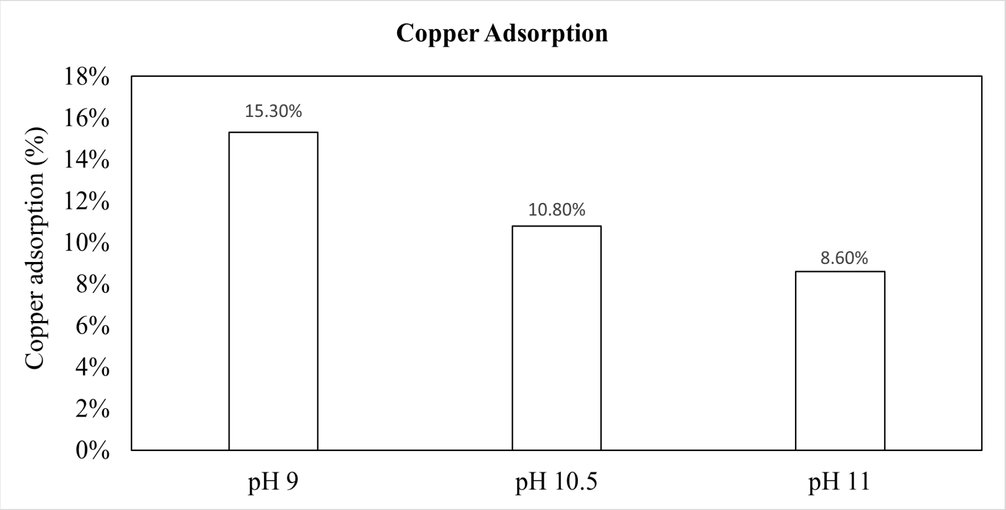

Figure 6. Percentage copper adsorption at different solution pH (Muchada H, et al.*, 2024).*

Figure 6 clearly shows that the highest copper adsorption was achieved in a pH 9 solution, which was because at low pH, copper di-cyanide Cu $(CN)_2^-$ was predominant in the solution, as previous research (Nicol *et al.*, 1987) reported that the formation of Cu $(CN)_2^-$ complex is highly favoured in a low pH. Copper adsorption strength was found to grow in the following order:

$$\mathrm{Cu(CN)_4^{3-}} < \mathrm{Cu(CN)_3^{2-}} < \mathrm{Cu(CN)_2^{-}}$$

Copper dicyanide Cu $(CN)_2^-$ was promptly and profoundly adsorbed by activated virgin carbon, providing high copper adsorption as illustrated in Figure 5.

Figure 6 shows a copper adsorption rate of 4.03×10^{-7} g/s over the first 15 minutes at pH 10.5. The initial steep copper concentration gradient further accelerated the adsorption rate. The amount of copper in the solution reduced gradually over the next 45 minutes to roughly 14.5 mg/L, yielding approximately 10.8% copper adsorption. The copper adsorption achieved in this particular solution was 4.5% less than that achieved in a pH 9 solution because at around pH 10 to 10.5 the concentration of Cu $(CN)_2^-$ substantially decreased in the solution and the presence of the copper tri-cyanide Cu $(CN)_3^{2-}$ was raised due to variations in solution pH.

Figure 6 shows that at pH 11, the copper adsorption rate was 3.77×10^{-7} g/s for the first 15 minutes, which was lower than the rates achieved in pH 9 and 10.5. Over the next 45 minutes, the amount of copper in the solution decreased moderately to around 14.86 mg/L, resulting in 8.6% copper adsorption. Copper adsorption in this solution was lower than at pH 9 and 10.5 by 6.7% and 2.2%, respectively. The pH 11 solution obtained the smallest copper adsorption, and this may operate as evidence to illustrate the stability of different types of cyanides created with copper with a change in solution pH. At pH 11, the presence of copper tetra-cyanide ($CN(CN)_4^{3-}$) rose substantially. The $CN(CN)_4^{3-}$ has the lowest adsorption strength. Based on the experimental results, pH 11 was determined to be the optimal pH for limiting excessive copper adsorption in the adsorption tanks.

Effect of pH on Gold Adsorption

Fig. 7. below shows the relationship between the concentration of gold in solution with increasing time.

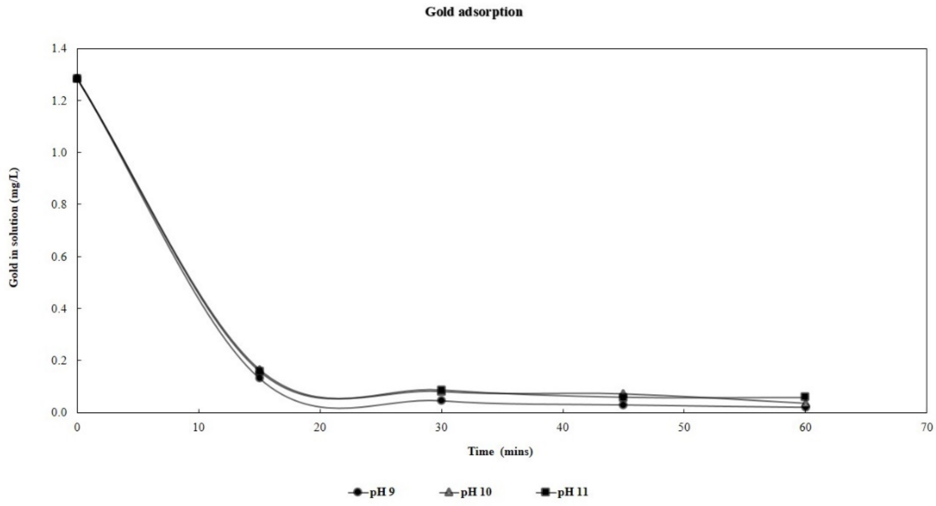

Figure 7. Gold concentration against time graph (initial gold assay = 1.29 mg/L, virgin carbon = 4 g and solution volume = 300 mL.

Generally, from the three curves the amount of gold in solution decreased from an initial value of 1.29 mg/L with increase in time. The three curves are comparably the same because in the first 15 minutes there was rapid gold adsorption. In further 45 minutes, the amount of gold in solution reached an almost constant level. However, considering 60 minutes pH 9, 10.5 and 11 solutions achieved gold adsorption of about 98.4%, 97.3% and 95.5%, respectively, meaning that increasing pH results in a decrease of gold adsorption. From the gold recoveries on the bar graph in Fig. 8, pH 9 yielded the highest copper and gold adsorption. Thus, pH 10 has fairly limited copper and gold adsorption and the solution pH should be maintained at pH 10.5 to avoid a significant decrease of gold adsorption whilst fairly limiting copper adsorption as well.

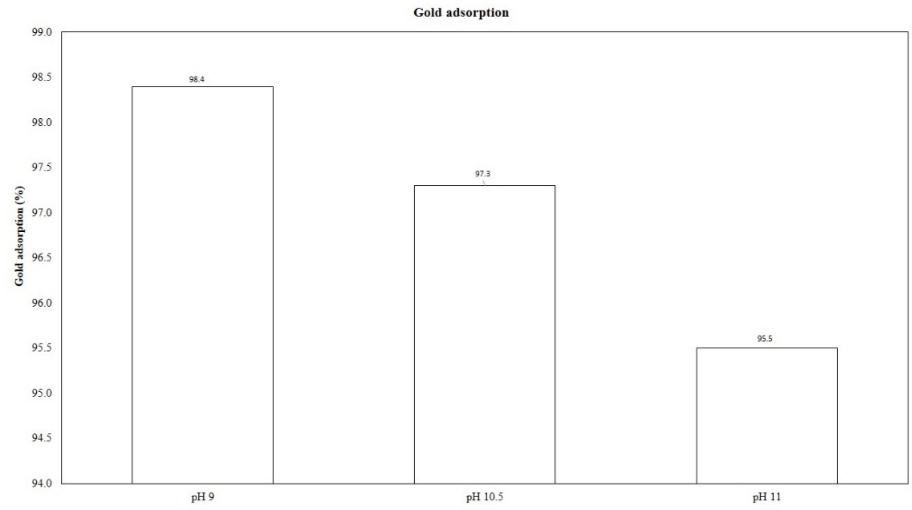

Figure 8. Percent gold adsorption with increasing solution pH.

Cold stripping - Cyanide Optimisation

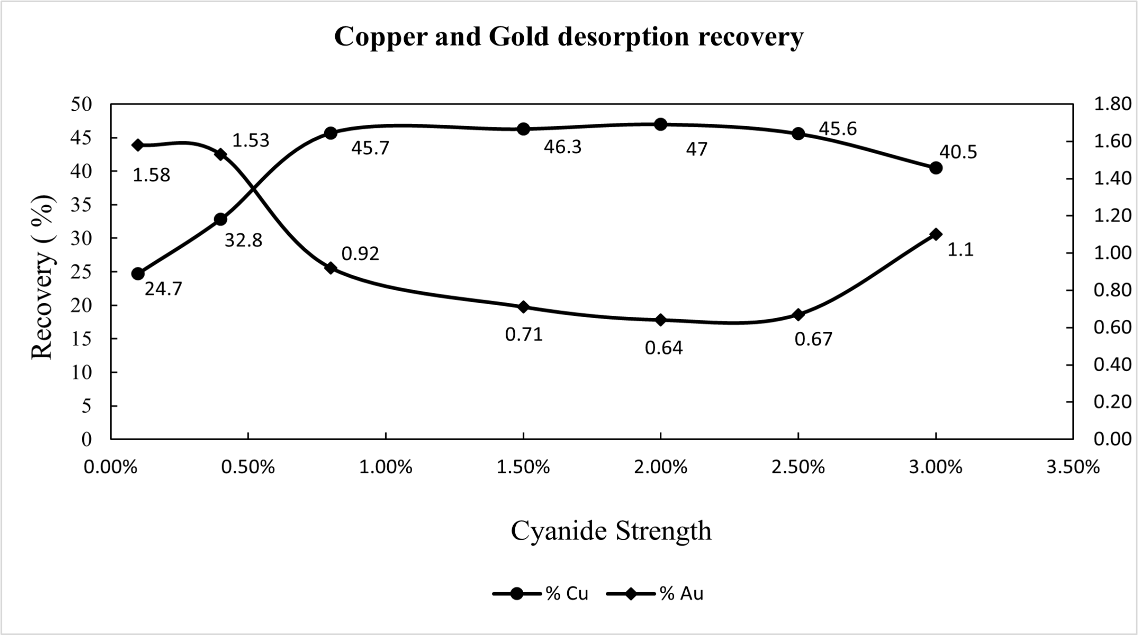

Figure 9. Copper and gold recovery with increasing cyanide strength from cold stripping experiments at [OH-]=1%, carbon mass=4 g, and solution volume=300 ml (Muchada H, et al.*, 2024)*

Figure 9 indicates the relationship of copper and gold recovery to cyanide strength. Increasing the cyanide strength from 0.1% to 2% resulted in an increase in copper desorption. However, increasing the cyanide strength from 2% to 3% resulted in decreased copper desorption. As a result, the highest copper desorption recovery was achieved at 2% cyanide strength. In contrast, when cyanide strength increased from 0.1% to 2%, gold desorption recovery reduced from 1.58% to 0.64%. The explanation for this declining tendency was that gold adsorption is highly advantageous at ambient temperature in the presence of copper; thus, increasing cyanide strength can improve competitive adsorption between gold and free cyanide ions. This indicates that as cyanide strength grows, the likelihood of gold in solution rapidly reaching equilibrium increases, and gold desorption recovery diminishes. However, from 2% to 3% cyanide, there was a moderate increase in gold desorption recovery from 0.64% to 1.1%, which was due to a decrease in copper desorption recovery, indicating that the solution becomes less saturated and the gold in solution equilibrium shifts upward, increasing gold desorption recovery. Figure 9 also reveals that maximum copper desorption recovery and minimum gold desorption recovery were attained at 2% cyanide strength, indicating that this was the ideal cyanide strength.

Effects of sodium hydroxide on copper desorption

Figure 10 depicts the fluctuation of copper over time at various sodium hydroxide concentrations during the selective copper stripping experiment.

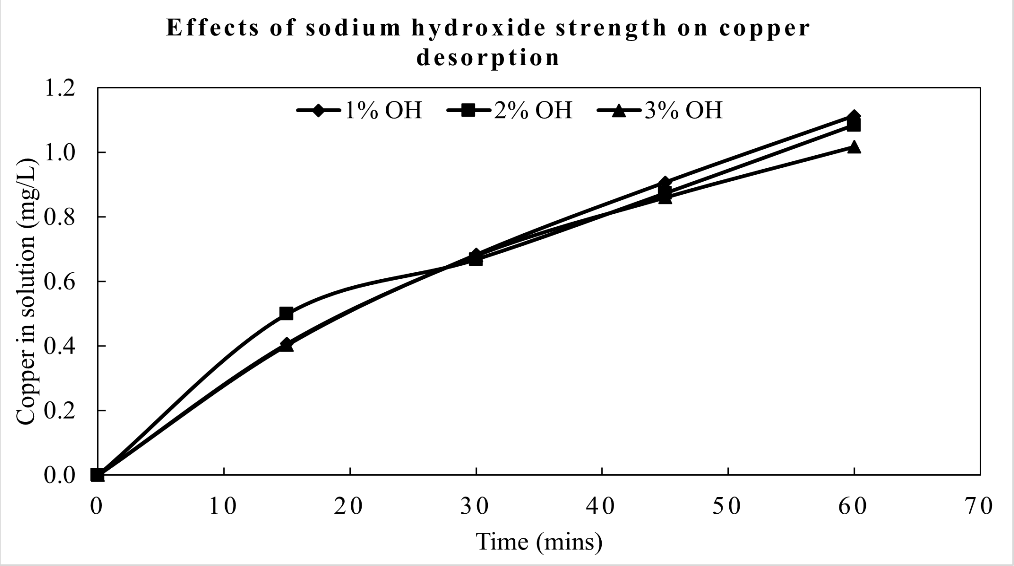

Figure 10. Copper in solution with increasing time for cold stripping test at [CN-]=0.4 %, carbon mass=4 %, and solution volume =300mL (Muchada H, et al.*, 2024).*

The data demonstrate that copper concentrations in solution increased rapidly, particularly in the first 10 minutes, indicating that copper complexes were weakly attracted to positively charged carbon adsorption sites. The three copper desorption curves in Figure 10 are nearly identical; however, around the 60-minute mark it is clear that the maximum quantity of copper in solution decreases with increasing sodium hydroxide strength. The sodium hydroxide strength was inversely proportional to copper desorption because increasing the sodium hydroxide strength increased pH, which increased the tendency of free cyanide to react with copper di-cyanide ($Cu\ (CN)_2^-$) and copper tri-cyanide ($Cu\ (CN)_3^{2-}$) to produce copper tetra-cyanide ($Cu\ (CN)_4^3$). As a result of Equations 8 and 9, the concentration of free cyanide ions in solution drops led to less copper desorption. The curves in Figure 10 show that the solution with 1% sodium hydroxide strength had a more efficient copper extraction than the other solutions since it had the greatest final concentration of copper in the solution, approximately 1.11 mg/L.

Figure 11 demonstrates that increasing the sodium hydroxide strength from 1% to 3% resulted in a net drop in copper desorption from 25.1% to 23%, indicating that 1% sodium hydroxide strength produced the best results.

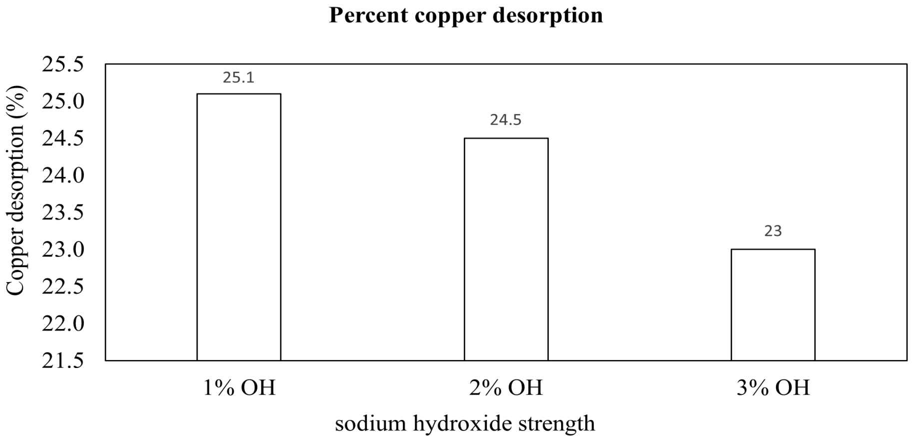

Figure 11. Percentage copper desorption with increasing sodium hydroxide strength during cold stripping at [CN-] = 0.4 %. Carbon mass = 4 g and solution volume = 300mL (Muchada H, et al.*, 2024).*

Effects of sodium hydroxide strength on gold desorption

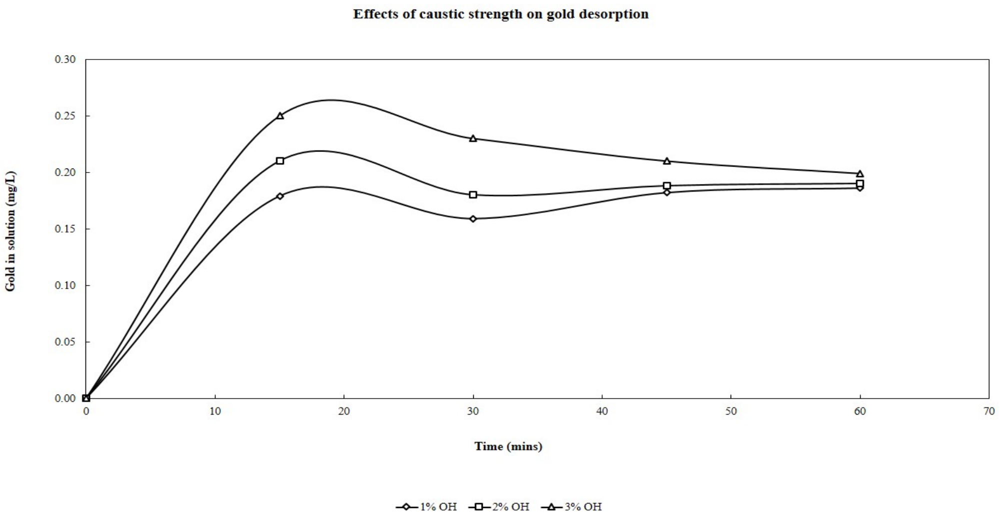

Figure 12. Gold desorption with increasing time at [CN-] = 0.4%, carbon mass = 4 g and solution volume = 300 mL.

The amount of gold that was desorbed from load activated carbon against time during copper cold stripping experiments containing varying sodium hydroxide strength is shown in Fig. 12. The desorption rate was rapid in the first 15 minutes of the experiment. This observation was due to the increase in the amount of hydroxyl ions in solution to assist in the displacement of gold from activated carbon adsorption sites (Yannopoulos, 1991). In a further 45 minutes, generally the curves were converging to their respective equilibrium level. At the 60-minute mark, an increase in sodium hydroxide resulted in an increase in gold in solution that could be attributed to the presence of hydroxyl ions in the solution. However, the amount of gold at the end of the three experiments was comparably equal (0.186, 0.190 and 0.199 mg/L). This was because a high amount of sodium hydroxide (high pH value) activates the reaction of the free cyanide ions with di- and trivalent copper complexes to form a predominate copper complex $(Cu (CN)_4^{3-}$, hence depleting the amount of free cyanide actively available for gold displacement. (Marsden & House, 2006).

The pie chart in Figure 13 shows that gold recovery increased with an increase in solution sodium hydroxide; therefore, 1% sodium hydroxide strength yielded the minimum gold recovery of 1.42%. Therefore, 1% sodium hydroxide was the most suitable for the cold stripping process.

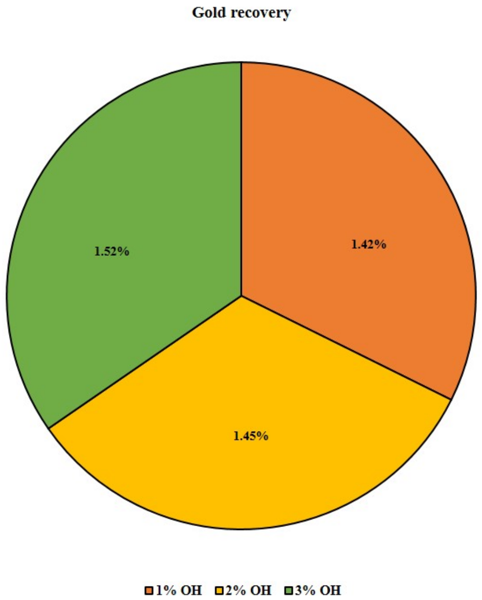

Figure 13. Percentage of gold recovery with increasing sodium hydroxide.

Effect of loaded carbon concentration on copper desorption

Figure 14 depicts the percent copper desorption achieved in cold stripping studies conducted at various carbon concentrations. Figure 14 shows that the percentage of copper extracted rose from 54.2% to 60% when the carbon content increased from 13 g/L to 27 g/L. This phenomenon results from increased activated carbon loading and contact area with the 300 mL solution. As a result, the desorption equilibrium promotes the mass transfer of copper cyanide complexes into solution via the macro and mesopores of activated carbon. However, increasing the carbon concentration reduced the percentage of copper extraction from 60% to 34.6%, indicating that the desorbed copper had saturated the 300 ml solution. As a result, the concentration of copper cyanide in the solution had achieved equilibrium. A carbon concentration of 27 g/L resulted in the maximum copper extraction of 60%; this was judged to be the best carbon concentration for a cold stripping experiment utilising a 300-mL solution containing 2% cyanide and 1% sodium hydroxide strength.

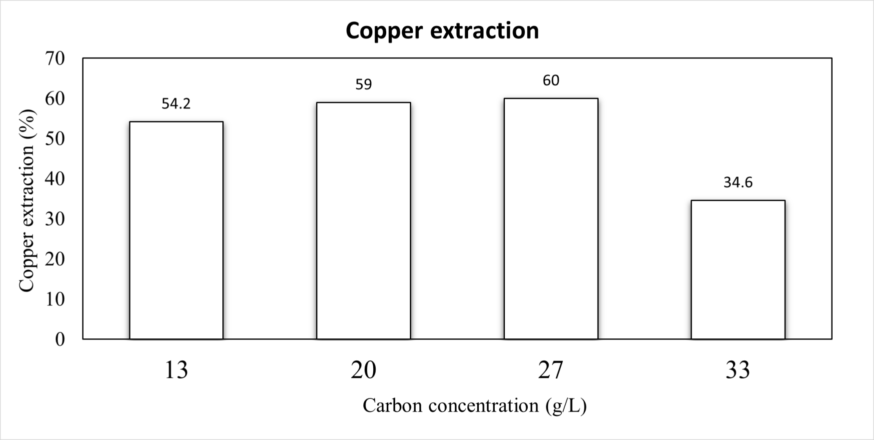

Figure 14. Percentage copper extracted with increasing carbon concentration at [CN-] =2 %, [OH-] = 1 % and solution volume = 300mL (Muchada H, et al.*, 2024).*

Effects of residence time on copper desorption
Figure 15 shows the percentage of copper extracted during the copper cold stripping experiment.

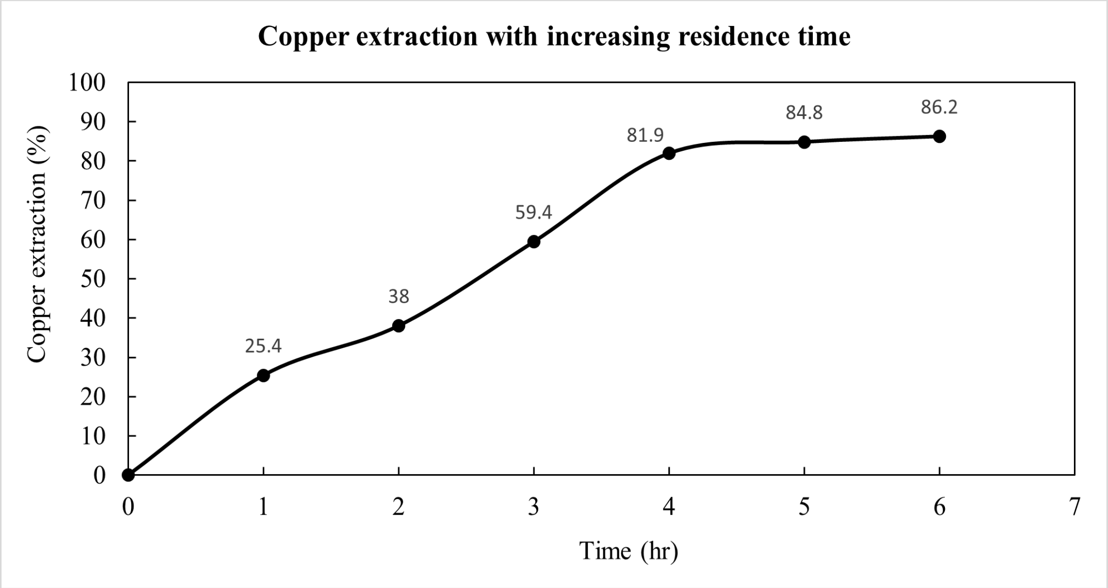

Figure 15, Percentage of copper extracted with increasing residence time (Muchada H, et al.*, 2024).*

Figure 15's curve demonstrated that as residence time grew, so did the percentage of copper removed. The percentage of copper removed grew until the curve reached a nearly constant level after four hours. This phenomenon might have been due to the copper cyanide in solution reaching equilibrium. It should also be noted that the increase in the percentage of copper removed was notably low - between four and six hours, indicating that an efficient copper extraction occurred for around four hours, making it the most acceptable cold stripping residence period in this study. At this point, the corresponding percent copper extraction was 81.9%, indicating that just 60 g/t of copper remained on the loaded carbon from the initial 332 g/t.

CONCLUSIONS

The following conclusions were drawn from this investigation:

1. The copper adsorption experiment showed that increasing the solution pH reduced copper adsorption. As a result, keeping the pH in the adsorption tanks at 10.5 would lower the adsorption of copper onto the activated carbon.
2. The optimum cyanide strength for the copper cold stripping experiment was 2%.

3. Increasing sodium hydroxide strength reduced copper desorption. As a result, 1% sodium hydroxide was found to be the best option since it produced the most copper desorption.
4. The effect of carbon concentration revealed that raising the carbon concentration tends to increase copper desorption up to an optimum point, after which additional increases have unfavourable effects. Thus, utilising 8g of loaded carbon in 300 ml (or 26.7 g/L) resulted in the highest copper recovery.
5. The influence of residence time enhanced the percentage of copper desorption. However, the desorption rate fell dramatically after four hours of residency. As a result, the most appropriate residence time for the copper cold stripping experiment was four hours.
6. The optimal conditions for decreasing copper in the solution were pH 10.5, 2% cyanide, 1% sodium hydroxide, 26.7 g/L carbon, and four hours of residence time. However, additional research was proposed to assess the desorption kinetics and execute a pilot plant trial to validate these findings.

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Assistant Lecturer in the Department of Materials Technology and Engineering at the Harare Institute of Technology and part time external assessor at Harare Polytechnic College. He holds a Bachelor's of Science Honours Degree in Metallurgical Engineering from University of Zimbabwe. Research areas include application of modern and economic mineral extraction technics in hydrometallurgy, pyrometallurgy and comminution of different ores.