

Gold Cementation from Ammonium Thiosulfate Solution by Zinc, Copper and Aluminium Powders

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Gold cementation test was conducted without de-aeration by using zinc, copper and aluminium powders from an ammonium thiosulfate solution contained 8 mg/l Au. The amount of metal powder was varied in the range of 30–450 Metal/Gold mass ratio. The solution composition was 1–5 mol/l NH₄OH, 0.01–0.05 mol/l CuSO₄ · 5H₂O, 0.2–0.4 mol/l (NH₄)₂S₂O₃ and pH 9.5–10.5. The results indicated that the gold was effectively recovered from a solution of lower ammonia and copper concentrations and higher thiosulfate concentration. The optimum reagent composition for the gold cementation from the ammonium thiosulfate solution was founded to be 1 mol/l NH₄OH, 0.01 mol/l CuSO₄ · 5H₂O and 0.4 mol/l (NH₄)₂S₂O₃ at pH 9.5. 100% of gold was recovered by zinc and aluminium powders at a Metal/Gold mass ratio of 30. Copper powder recovered 93% of gold at a Metal/Gold mass ratio of 50. Zinc might re-generate thiosulfate concentration and precipitate most of copper in the solution. Aluminium precipitation might recover gold with less amount of copper deposition and some thiosulfate reduction. Copper precipitation reduced a small amount of thiosulfate concentration and greatly increased copper concentration. Ammonia concentration stayed constant during cementation process.

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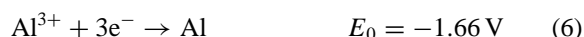
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1. Introduction

Ammonium thiosulfate leaching is an alternative gold extraction method to cyanidation because of its low toxicity.^{1–7)} However, the gold recovery method from the leach solution was not widely studied. In our study of gold leaching,⁸⁾ an optimum condition to extract gold from Hishikari ore was developed. In this paper, the cementation method to recover gold from the leach solution is presented and discussed.

CIP or CIL and Merrill Crowe processes are the standard gold recovery process in cyanidation.^{9–14)} It was reported that aurous thiosulfate complex has no affinity to activated carbon.¹⁵⁾ Therefore, CIP or CIL processes is not applicable in the ammonium thiosulfate leaching process. Marchbank *et al.* suggested the addition of cyanide to thiosulfate solution followed by carbon adsorption.¹⁶⁾ However, this method cannot be applied in the wholly non-cyanide process.

Gold cementation by metal powders may be considered as one of the best gold recovery methods. The use of zinc, copper and aluminium could be the candidate for gold cementation because of their lower reduction potentials than gold as shown in eqs. (1)–(6).¹⁷⁾



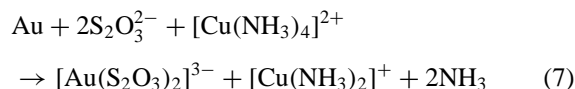
The use of zinc, copper and aluminium has been suggested

to recover gold from thiosulfate solution,^{18–20)} but the effect of copper, thiosulfate and ammonia concentration on the required amount of metal powder was not reported. The objective of this investigation was to find the relationship between the solution composition and the amount of metal powder required, and to establish the optimum condition for gold cementation.

2. Chemistry

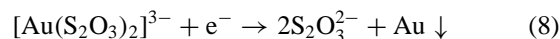
2.1 Leach solution

In the ammonium thiosulfate solution, gold and copper occurred in the form of amine or thiosulfate complexes, such as [Au(NH₃)]⁺, [Cu(NH₃)₄]²⁺, [Cu(NH₃)₂]⁺, [Au(S₂O₃)₂]³⁻ and [Cu(S₂O₃)₂]³⁻. According to the reaction chemistry of gold leaching as shown in eq. (7),^{5–8)} it is assumed that most of copper should form amine complex, and gold should form thiosulfate complex:

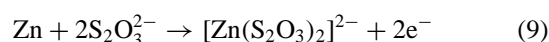


2.2 Zinc precipitation

The main cathodic reaction of gold cementation by zinc powder in the ammonium thiosulfate solution may be expressed in eq. (8):

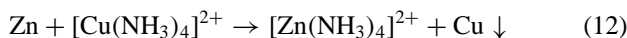
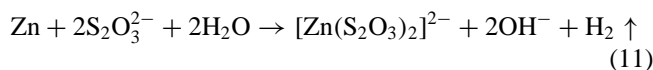
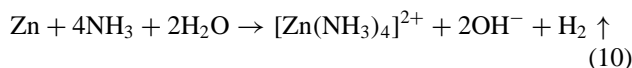


The anodic reaction of zinc precipitation is shown in eq. (9).^{21–24)}

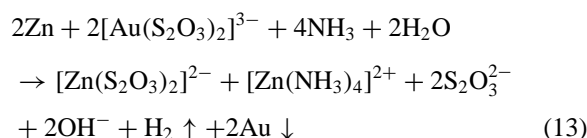


Since the pregnant solution contained ammonia, thiosulfate and cupric amine complex, following reactions may also oc-

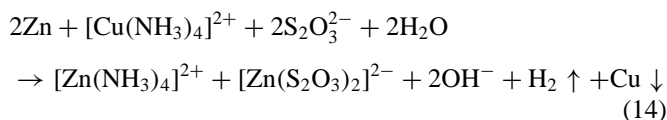
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cur:^{21–24)}

The combination of eqs. (8), (9) and (10) is the overall reaction of gold cementation by zinc powder in ammonium thiosulfate solution:



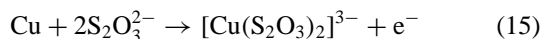
Equation (13) indicates that one mole of zinc can precipitate one mole of gold. Also, copper precipitation by zinc powder could be occurred in the sum of eqs. (11) and (12):



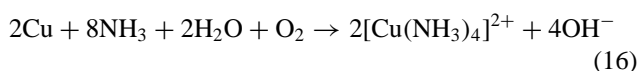
Equation (14) indicates that two moles of zinc could precipitate one mole of copper in the solution.

2.3 Copper precipitation

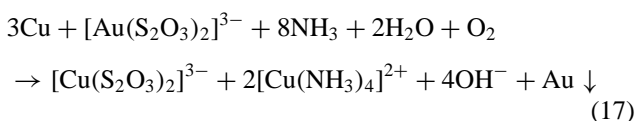
In gold cementation by copper powder, cathodic reaction (8) will take place along with the following anodic reaction (15):^{19,23)}



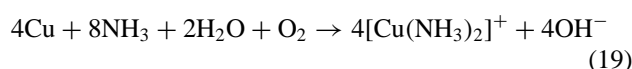
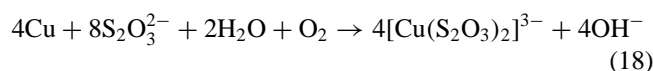
Also, copper powder will be oxidized to cupric amine complex by ammonia as shown in eq. (16):^{21–24)}



The combination of eqs. (8), (15) and (16) is the overall reaction of gold cementation with copper:

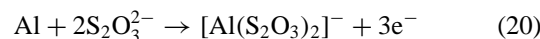


Equation (17) demonstrates that one mole of gold needs three moles of copper in copper precipitation. Also, more copper powder could be oxidized as cuprous amine and thiosulfate complexes as shown in eqs. (18) and (19):

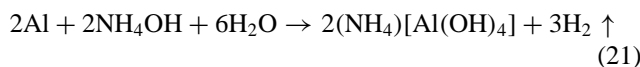


2.4 Aluminium precipitation

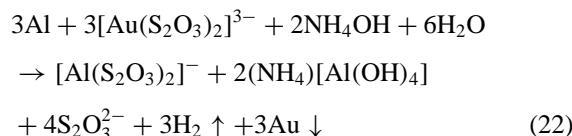
In gold cementation by aluminium powder, cathodic reaction (8) will take place along with the anodic reaction

(20).^{18,23)}

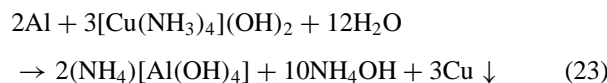
Also, aluminium will be oxidized in the ammonia solution into aluminium hydroxo complex as shown in eq. (21):^{21,24)}



The combination of eqs. (8), (20) and (21) is the overall reaction of gold cementation with aluminium:



Equation (22) indicates that one mole of gold can be precipitated by one mole of aluminium. Also, cupric amine complex in the solution could be reduced by aluminium as shown in eq. (23):



Thus, the copper content in the barren solution could be diminished slightly:^{21,22)}

3. Experimental

Table 1 shows the ammonium thiosulfate leaching conditions and the final compositions of the pregnant solutions obtained from a silicate type Hishikari ore.⁸⁾ The range of reagent concentration were 1–5 mol/l NH_4OH , 0.01–0.05 mol/l CuSO_4 and 0.2–0.4 mol/l, $(\text{NH}_4)_2\text{S}_2\text{O}_3$ and pH 9.5–10.5. The pH was adjusted by adding 10 vol% H_2SO_4 or NaOH solution. The gold concentration in the pregnant solution was 8 mg/l Au.

Average particle size of metal powder used for the gold cementation was 45 μm . The amount of metal powders required was calculated and added in a 100 ml pregnant solution. The sample was gently agitated by a magnetic stirrer for 60 min at ambient temperature without de-aeration.

After the gold cementation, the pulp was filtered with micro filter immediately. The filtrate was assayed for gold, cop-

Table 1 Composition of the Initial and the Pregnant Solutions.⁸⁾

NH_4OH (mol/l)		CuSO_4 (mol/l)		$(\text{NH}_4)_2\text{S}_2\text{O}_3$ (mol/l)		pH		Au (mg/l)
Initial	Preg.	Initial	Preg.	Initial	Preg.	Initial	Preg.	Preg.
3	3	0.03	0.026	0.3	0.18	10.0	10.0	8.17
1	1	0.03	0.027	0.3	0.16	9.5	9.5	8.41
5	5	0.03	0.026	0.3	0.19	10.2	10.2	8.43
3	3	0.01	0.007	0.3	0.23	10.0	10.0	8.05
3	3	0.05	0.043	0.3	0.18	10.0	10.0	8.18
3	3	0.03	0.026	0.2	0.10	10.0	10.0	8.13
3	3	0.03	0.027	0.4	0.28	10.0	10.0	8.21
3	3	0.03	0.026	0.3	0.24	9.5	9.5	8.40
3	3	0.03	0.022	0.3	0.17	10.5	10.5	8.18
1	1	0.01	0.008	0.4	0.33	9.5	9.5	8.37

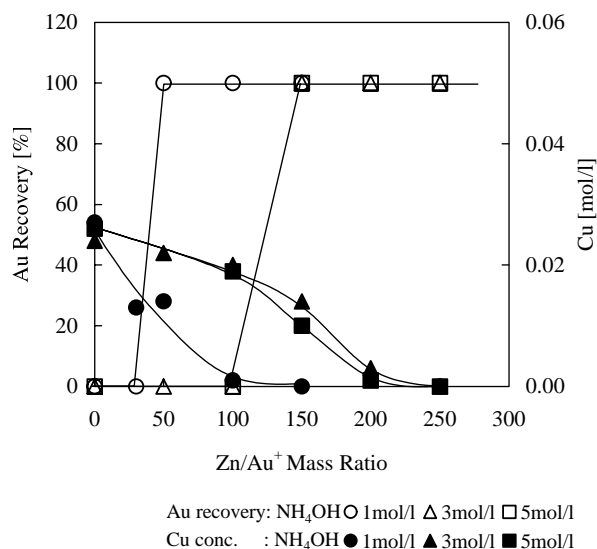


Fig. 1 Effect of Zn/Au^+ mass ratio and ammonia concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5–10.2, 60 min agitation.

per, thiosulfate and ammonia. Gold and copper concentrations were measured by atomic adsorption spectrophotometer. The thiosulfate concentration was measured by idiometric titration.²⁵⁾ The ammonia concentration was measured by Nessler's method.²⁶⁾ The sensitivity of Nessler's method is at semi-quantitative level, and the standard deviation is around $\pm 0.5 \text{ mol/l } \text{NH}_4^+$. The effects of ammonia, copper and thiosulfate concentrations and pH on the gold recovery were investigated. To investigate the feasibility of recycling the ammonium thiosulfate solution, the total reductions of ammonia, copper and thiosulfate due to the gold cementation process were evaluated.

4. Result and Discussion

4.1 Effect of ammonia and metal/gold mass ratio

Figure 1 shows the effect of Zn/Au^+ mass ratio and ammonia concentration on gold recovery and final copper concentration. The results show that the optimum Zn/Au^+ mass ratio to recover 100% of gold in the presence of 1, 3 and 5 mol/l NH_4OH were 50, 150 and 150 respectively. The final copper concentrations at the optimum Zn/Au^+ mass ratio in the 1, 3 and 5 mol/l NH_4OH solutions were 0.010, 0.014 and 0.010 mol/l respectively. It may be concluded that larger amount of zinc powder was required at higher ammonia concentration to recover gold. The cause of zinc consumption may be due to the oxidation reaction and the copper precipitation as shown in eqs. (10), (11) and (12). Thus, low ammonia concentration could minimize the required zinc powder for gold recovery.

Figure 2 shows the effect of Cu/Au^+ mass ratio and ammonia concentration on gold recovery and final copper concentration. The optimum Cu/Au^+ mass ratio to recover more than 90% of gold in the 1, 3 and 5 mol/l NH_4OH solutions were 200, 350 and 400 respectively. The maximum gold recovery could not achieve 100% but 95%. The results may be due to the re-dissolution of precipitated gold by the cat-

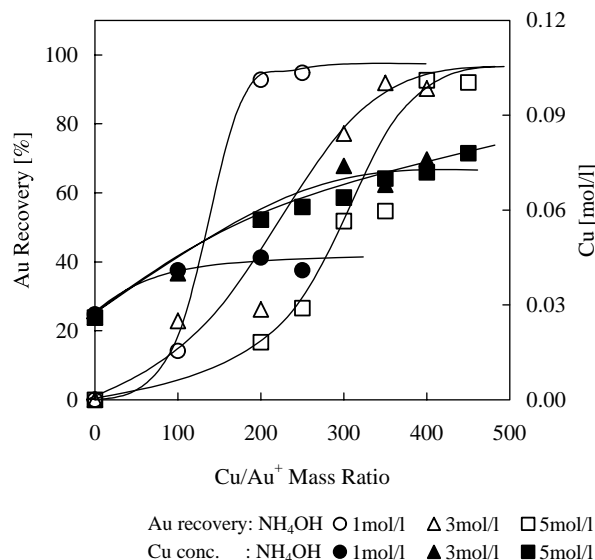
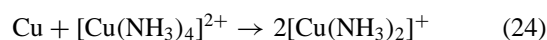
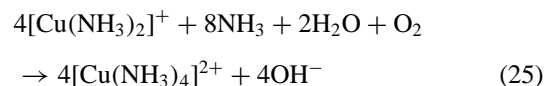


Fig. 2 Effect of Cu/Au^+ mass ratio and ammonia concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5–10.2, 60 min agitation.

alytic action of cupric amine complex as shown in eq. (7). The final copper concentrations at the optimum Cu/Au^+ mass ratio in the 1, 3 and 5 mol/l NH_4OH solutions were 0.045, 0.068 and 0.072 mol/l respectively. Additional 0.015, 0.038 and 0.042 mol/l of copper was dissolved in the solution. The large consumption of copper powder at the higher ammonia concentration may be attributed to the formation of cuprous amine and thiosulfate complexes as shown in eqs. (18) and (19). During the gold cementation, the dark blue solution was gradually faded away to light blue. The evidence demonstrates that cupric amine complex was reduced to cuprous amine complex by the copper powder dissolution as shown in eq. (24):¹⁹⁾



When the light blue-barren solution was left for a couple of hours, the colour was back to dark blue gradually. This fact indicates that the large amount of cuprous amine complex was gradually oxidized to cupric amine complex as shown in eq. (25):^{8,19)}



The results may be concluded that a lower ammonia concentration is required to recover the gold with smaller copper powder consumption.

Figure 3 shows the effect of Al/Au^+ mass ratio and ammonia concentration on gold recovery and final copper concentration. The minimum Al/Au^+ mass ratio to recover more than 90% of gold in the 1, 3 and 5 mol/l NH_4OH solutions were 100, 350 and 150 respectively. The final copper concentrations at the optimum Al/Au^+ mass ratio in the 1, 3 and 5 mol/l NH_4OH solutions were 0.025, 0.022 and 0.022 mol/l respectively. Less than 0.005 mol/l of copper was co-precipitated with gold. The 3 mol/l NH_4OH solution consumed a larger amount of aluminium powder than the

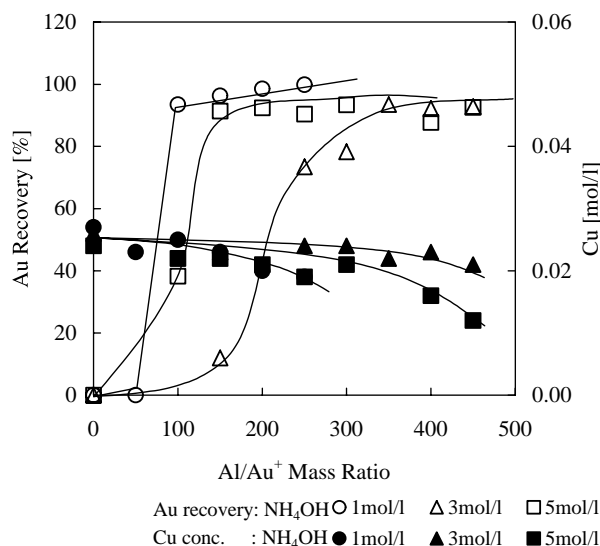


Fig. 3 Effect of Al/Au^+ mass ratio and ammonia concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5–10.2, 60 min agitation.

solutions of 1 and 5 mol/l NH_4OH . This may be explained that 1 mol/l of ammonia was not enough to stabilize copper ion as cupric amine complex. In our previous study,⁸⁾ the thiosulfate consumption was at lowest between 3 and 5 mol/l NH_4OH . When the NH_4OH concentration decreased to 1 mol/l, the thiosulfate consumption increased. Therefore, it is assumed that the stability of $[\text{Cu}(\text{NH}_3)_4]^{2+}$, which is the catalyst for gold extraction, was greatly increased with the increase of ammonia concentration from 1 mol/l to 3 mol/l, and the Al/Au^+ mass ratio required was increased. Further increase of NH_4OH to 5 mol/l, the stability of $[\text{Al}(\text{OH})_4]^-$ became higher than that of $[\text{Cu}(\text{NH}_3)_4]^{2+}$ due to the increase of OH^- and reduced the Al/Au^+ mass ratio required. The consumption of aluminium may be due to the production of $[\text{Al}(\text{OH})_4]^-$ as shown in the eq. (21) and the formation of $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ (boehmite) as shown in eq. (26).²²⁾



The copper content was slightly diminished at 0.005–0.008 mol/l. The results could be due to the production of aluminium hydroxo complex by copper reduction as explained in eq. (23).

4.2 Effect of copper sulphate and metal/gold mass ratio

Figure 4 shows the effect of Zn/Au^+ mass ratio and copper sulphate concentration on gold recovery and final copper concentration. The optimum Zn/Au^+ mass ratio to recover 100% of gold in the 0.01, 0.03 and 0.05 mol/l CuSO_4 solutions were 30, 150 and 250 respectively. The correspondent final copper concentrations at the optimum Zn/Au^+ mass ratio in the 0.01, 0.03 and 0.05 mol/l CuSO_4 solutions were 0.003, 0.014 and 0.023 mol/l. The total copper loss at the optimum Zn/Au^+ mass ratio in the 0.01, 0.03 and 0.05 mol/l CuSO_4 solutions were 0.007, 0.016 and 0.027 mol/l, which indicated that about half of copper was precipitated from the initial concentration. The precipitation of copper may be attributed to the co-precipitation with gold by zinc as shown in eq. (14).

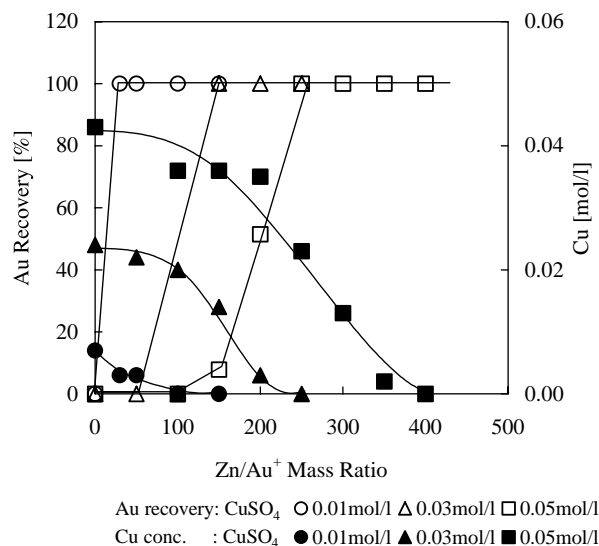


Fig. 4 Effect of Zn/Au^+ mass ratio and copper sulphate concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH_4OH and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 10.0, 60 min agitation.

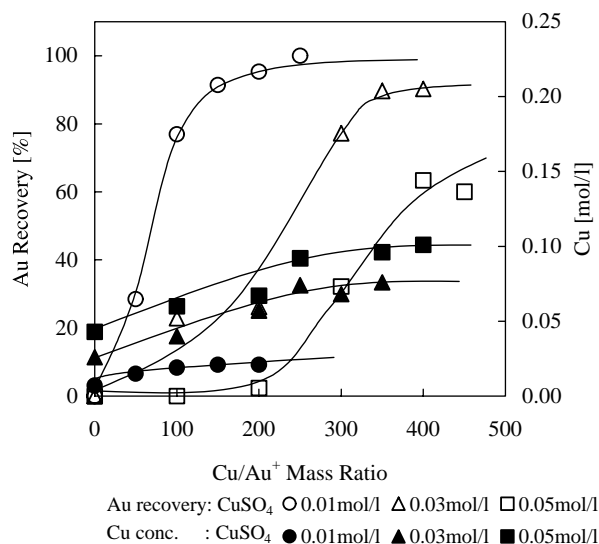


Fig. 5 Effect of Cu/Au^+ mass ratio and copper sulphate concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH_4OH and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 10.0, 60 min agitation.

Thus, a low copper concentration could result in a high gold recovery with a low copper powder consumption.

Figure 5 shows the effect of Cu/Au^+ mass ratio and copper sulphate concentration on gold recovery and final copper concentration. The optimum Cu/Au^+ mass ratio to recover more than 90% of gold in the 0.01 and 0.03 mol/l CuSO_4 solutions were 150 and 350 respectively. The final copper concentrations at the optimum Cu/Au^+ mass ratio in the 0.01 and 0.03 mol/l CuSO_4 solutions were 0.021 and 0.068 mol/l respectively. The amount of dissolved copper concentration was double. The 0.05 mol/l CuSO_4 solution could not recover gold more than 65% in the range of 0 to 450 Cu/Au^+ mass ratio. The results indicate that high copper sulphate concentration could require more copper powder. The large consumption of copper powder at high copper sulphate concentration may be attributed to the production of cuprous thiosulfate and amine complexes as shown in eqs. (18), (19) and (24). In or-

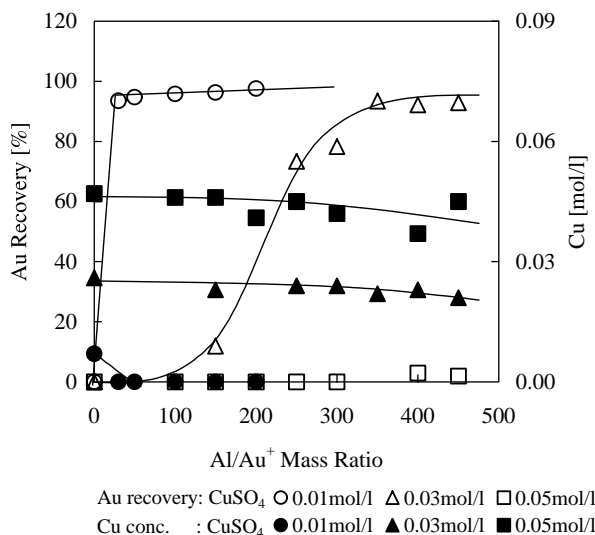


Fig. 6 Effect of Al/Au⁺ mass ratio and copper sulphate concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH₄OH and 0.3 mol/l (NH₄)₂S₂O₃ at pH 10.0, 60 min agitation.

der to minimize the copper powder consumption, it is better to minimize the initial copper sulphate concentration at both leaching and cementation stages.

Figure 6 shows the effect of Al/Au⁺ mass ratio and copper sulphate concentration on gold recovery and final copper concentration. The optimum Al/Au⁺ mass ratio to recover more than 90% of gold in the 0.01 and 0.03 mol/l CuSO₄ solutions were 30 and 350 respectively. The final copper concentrations at the optimum Al/Au⁺ mass ratio in the 0.01 and 0.03 mol/l CuSO₄ solutions were 0.000 and 0.022 mol/l respectively. The 0.05 mol/l CuSO₄ solution could not recover gold in the range of 0 to 450 Al/Au⁺ mass ratio. As the copper concentration increased, the more aluminium powder was consumed. The aluminium consumption may be attributed to the oxidation reaction of Al to [Al(OH)₄]⁻ or Al₂O₃ · H₂O as indicated in eqs. (19) and (24). Thus, a low copper concentration is preferred for minimizing the consumption of aluminium powder.

4.3 Effect of ammonium thiosulfate and metal/gold mass ratio

Figure 7 shows the effect of Zn/Au⁺ mass ratio and ammonium thiosulfate concentration on gold recovery and final copper concentration. The optimum Zn/Au⁺ mass ratio to recover 100% of gold in the 0.2, 0.3 and 0.4 mol/l (NH₄)₂S₂O₃ solutions were 200, 150, 100 respectively. The correspondent final copper concentrations at the optimum Zn/Au⁺ mass ratio in the 0.2, 0.3 and 0.4 mol/l (NH₄)₂S₂O₃ solutions were 0.011, 0.014 and 0.016 mol/l respectively. Higher thiosulfate concentration reduced both the zinc powder required and the loss of copper ion in gold recovery. The results indicate that zinc powder was oxidized easily as [Zn(S₂O₃)₂]²⁻ with reducing [Au(S₂O₃)₂]³⁻ at a higher thiosulfate concentration. It also confirms the results of Fig. 1, which indicated that the lower ammonia concentration recovered more gold at the lower Zn/Au⁺ mass ratio due to the increase of S₂O₃²⁻/NH₃(aq) mole ratio. Thus, higher thiosulfate concentration would minimize the amount of zinc powder required.

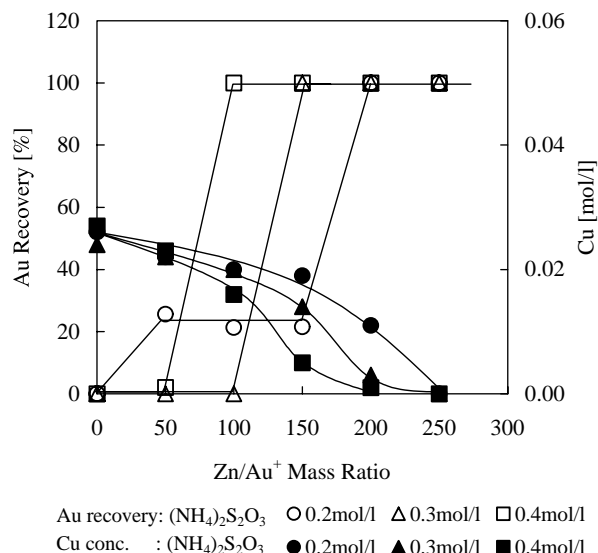


Fig. 7 Effect of Zn/Au⁺ mass ratio and ammonium thiosulfate concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH₄OH and 0.03 mol/l CuSO₄ at pH 10.0, 60 min agitation.

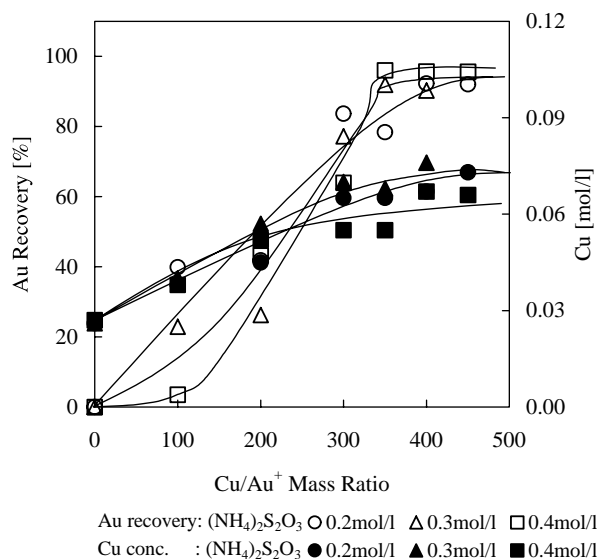


Fig. 8 Effect of Cu/Au⁺ mass ratio and ammonium thiosulfate concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH₄OH and 0.03 mol/l CuSO₄ at pH 10.0, 60 min agitation.

Figure 8 shows the effect of Cu/Au⁺ mass ratio and thiosulfate concentration on gold recovery and final copper concentration. The optimum Cu/Au⁺ mass ratio to recover more than 90% of gold in the 0.2, 0.3 and 0.4 mol/l (NH₄)₂S₂O₃ solutions were 400, 350, 350 respectively. The final copper concentrations at the optimum Cu/Au⁺ mass ratio in the 0.2, 0.3 and 0.4 mol/l (NH₄)₂S₂O₃ solutions were 0.067, 0.068 and 0.055 mol/l respectively. The large amount of copper powder was required at the low thiosulfate concentration. The result suggests that copper powder was oxidized to [Cu(S₂O₃)₂]³⁻ at a higher thiosulfate concentration. Thus, the condition of high thiosulfate concentration could result in a high gold recovery with a lower copper consumption.

Figure 9 shows the effect of Al/Au⁺ mass ratio and ammonium thiosulfate concentration on gold recovery and final

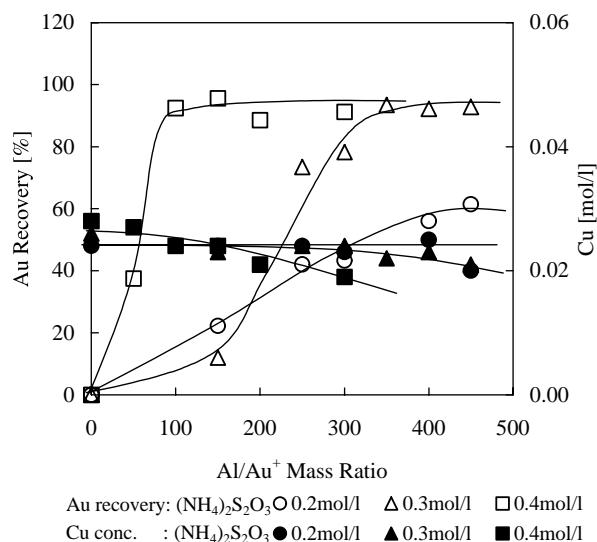
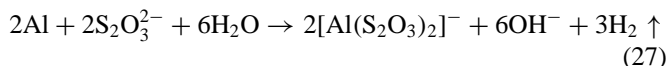


Fig. 9 Effect of Al/Au^+ mass ratio and ammonium thiosulfate concentration on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH_4OH and 0.03 mol/l CuSO_4 at pH 10.0, 60 min agitation.

copper concentration. The maximum gold recovery in the 0.2 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ solution was 61% at 450 of Al/Au^+ mass ratio. The optimum Al/Au^+ mass ratio to recover more than 90% of gold in the 0.3 and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ solutions were 350 and 100 respectively. The final copper concentrations at the optimum Al/Au^+ mass ratio in the 0.3 and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ solutions were 0.022 and 0.024 mol/l. The results indicate that thiosulfate may oxidize aluminium as a product of $[\text{Al}(\text{S}_2\text{O}_3)_2]^-$ as shown in eq. (27):



Compared to the result of zinc precipitation as shown in Fig. 7, the diminished copper concentration was low at around 0.007 mol/l, which support the results that aluminium was oxidized to thiosulfate complex rather than amine complex. In conclusion, 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ should be the optimum concentration for the gold recovery by aluminium precipitation.

4.4 Effect of pH and Metal/Gold mass ratio

Figure 10 shows the effect of Zn/Au^+ mass ratio and pH on gold recovery and final copper concentration. At the reagent combination of 3 mol/l NH_4OH , 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$, the optimum Zn/Au^+ mass ratio to recover 100% of gold was 150 at the pH range of 9.5–10.5. The copper concentrations at the optimum Zn/Au^+ mass ratio in the pH 9.5, 10.0 and 10.5 solutions were 0.021, 0.014 and 0.016 mol/l respectively. The result indicates that lower pH might prevent the copper precipitation. The cause of lower copper consumption at pH 9.5 is assumed that cupric amine worked as an oxidant for zinc powder to produce zinc thiosulfate complex and assisted to reduce aurous thiosulfate complex as shown in eq. (28):

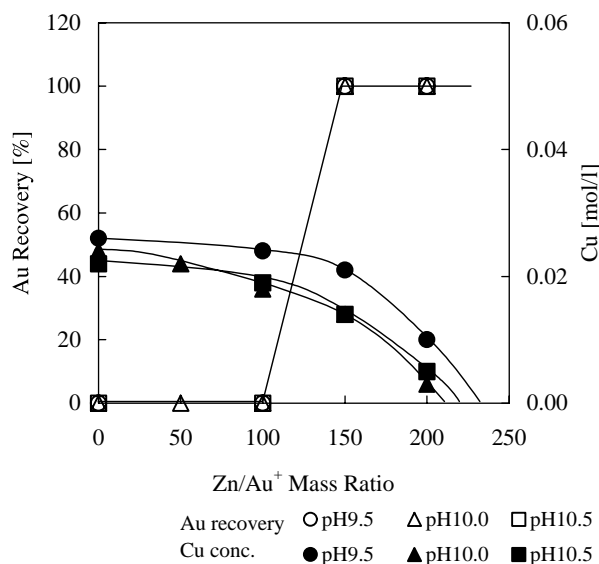
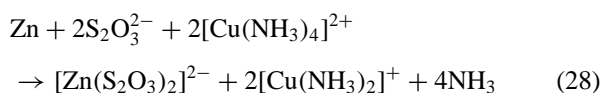


Fig. 10 Effect of Zn/Au^+ mass ratio and pH on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH_4OH , 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$, 60 min agitation.

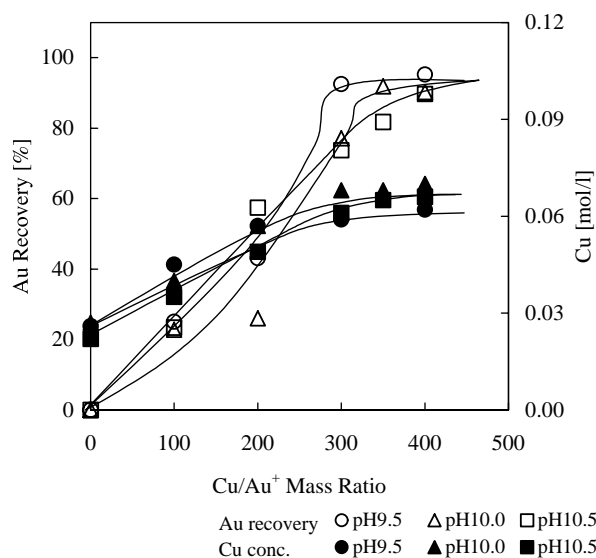


Fig. 11 Effect of Cu/Au^+ mass ratio and pH on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH_4OH , 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$, 60 min agitation.

Copper ion is stable as amine complex rather than thiosulfate complex around pH 9.5,^{8,27)} and zinc has an affinity for both ammonia and thiosulfate to form $[\text{Zn}(\text{NH}_3)_4]^{2+}$ and $[\text{Zn}(\text{S}_2\text{O}_3)_2]^{2-}$ as explained in eqs. (10) and (11). This evidence also supports the eq. (28). Thus, pH 9.5 could be the optimum pH for zinc precipitation.

Figure 11 shows the effect of Cu/Au^+ mass ratio and pH on gold recovery and final copper concentration. At the reagent combination of 3 mol/l NH_4OH , 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$, the optimum Cu/Au^+ mass ratio to recover 90% of gold in the pH 9.5, 10.0 and 10.5 solutions were 300, 350 and 400 respectively. The copper concentrations at the optimum Cu/Au^+ mass ratio in the solutions at pH 9.5, 10.0 and 10.5 were 0.059, 0.068 and 0.066 mol/l respectively. The lower the pH was, the less the copper was consumed. The small amount of copper powder required at

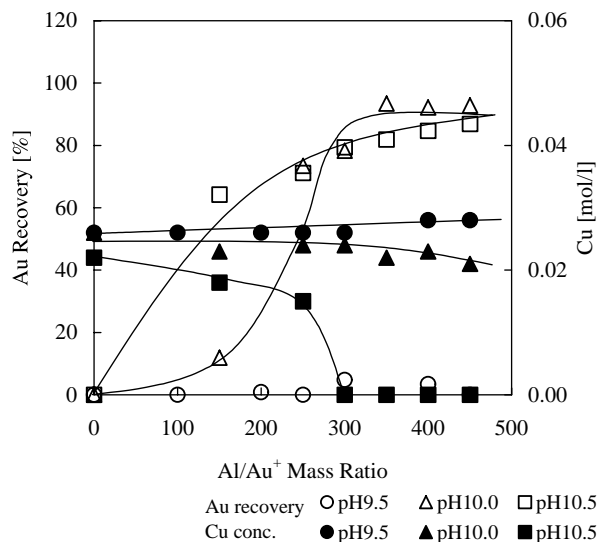
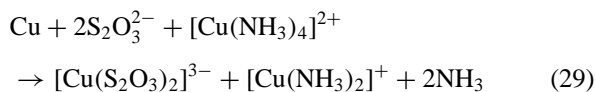


Fig. 12 Effect of Al/Au^+ mass ratio and pH on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 3 mol/l NH_4OH , 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$, 60 min agitation.

the pH 9.5 may be attributed to the activation of cupric amine complex as the oxidant for copper powder to produce cuprous thiosulfate complex. In the study of gold leaching,⁸⁾ it was confirmed that Cu^{2+} is stable as $[\text{Cu}(\text{NH}_3)_4]^{2+}$ rather than CuO at pH around 9.5. These facts suggest that copper powder was oxidized to $[\text{Cu}(\text{S}_2\text{O}_3)_2]^{3-}$ readily by the catalytic action of $[\text{Cu}(\text{NH}_3)_4]^{2+}$ as pH decreased from 10.5 to 9.5 in eq. (29):



Thus, pH 9.5 could be the optimum condition to cement more gold with less copper powder consumption.

Figure 12 shows the effect of Al/Au^+ mass ratio and pH on gold recovery and final copper concentration at the reagent combination of 3 mol/l NH_4OH , 0.03 mol/l CuSO_4 and 0.3 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$. The results showed that the solution at pH 9.5 could not recover gold by aluminium in the range of 0 to 450 Al/Au^+ mass ratio. The optimum Al/Au^+ mass ratio to recover more than 80% of gold in the solutions at pH 10.0 and 10.5 was 350 for both cases. The final copper concentrations at the optimum Al/Au^+ mass ratio in the solutions at pH 10.0 and 10.5 were 0.022 and 0.000 mol/l respectively. The solution at pH 10.5 precipitated entire copper in the ammonium thiosulfate solution. The large copper consumption may be attributed to the increase of aluminium solubility at high pH. Aluminium cannot form amine complex. It means that $[\text{Cu}(\text{NH}_3)_2]^{2+}$ cannot work as an oxidant for aluminium oxidation to form thiosulfate complex like Au, Zn and Cu as shown in eqs. (7), (28) and (29). Aluminium is less soluble as $[\text{Al}(\text{OH})_4]^-$ at a lower pH.^{21,22)} These facts suggest that the increase of pH without the use of ammonia causes the large copper consumption due to the increase of the $[\text{Al}(\text{OH})_4]^-$ stability against the decrease of the $[\text{Cu}(\text{NH}_3)_4]^{2+}$ stability as explained in eq. (23). The poor gold recovery at pH 9.5 could be attributed to the increase of $[\text{Cu}(\text{NH}_3)_4]^{2+}$ stability and the poor $[\text{Al}(\text{OH})_4]^-$ stability.

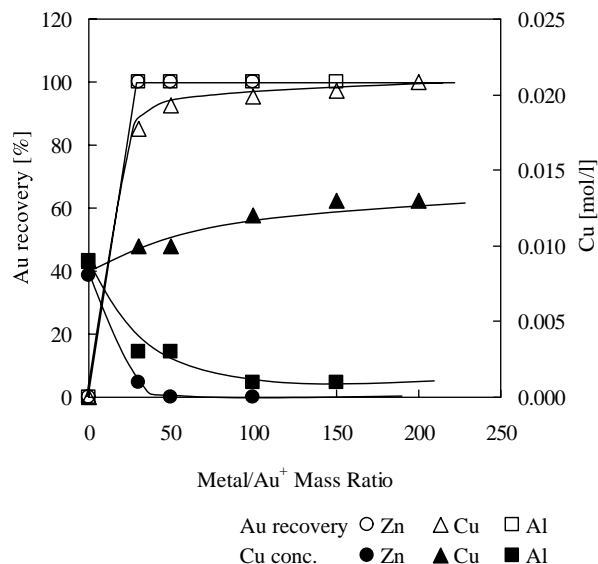


Fig. 13 Effect of Metal/Au^+ mass ratio on gold recovery and copper concentration. 8 mg/l-Au pregnant solution with 1 mol/l NH_4OH and 0.01 mol/l CuSO_4 and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5, 60 min agitation.

The pH for the optimum combination at 0.01 mol/l CuSO_4 , 1 mol/l NH_4OH and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ is 9.5. At this lower ammonia and copper concentrations and higher thiosulfate concentration, pH adjustment would not be required for gold cementation.

4.5 Final evaluation for Zn, Cu and Al precipitation

4.5.1 Gold recovery and copper concentration

From the results of Figs. 1–12, the optimum condition could be concluded as 1 mol/l NH_4OH , 0.01 mol/l CuSO_4 and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5. Figure 13 confirms the effect of Metal/Gold mass ratio on gold recovery and final copper concentration at this optimum condition. The optimum Zn/Au^+ and Al/Au^+ mass ratio to recover 100% of gold were 30. The optimum Cu/Au^+ mass ratio to recover 90% of gold was 50. To achieve 100% of gold recovery by copper powder, a Cu/Au^+ mass ratio of 200 was required. The final copper concentrations at the optimum Metal/Gold mass ratio for Zn, Cu and Al were 0.001, 0.009 and 0.003 mol/l respectively. Copper is not only the most expensive metal among the used metal powders but also required a larger Metal/ Au^+ mass ratio than that of zinc and aluminium. The total reduction of copper concentration by zinc and aluminium powders was respectively 0.009 mol/l and 0.007 mol/l. The replenished amount of copper in zinc precipitation is more than that in aluminium precipitation process. The excess addition of copper powder increased the copper dissolution. For example, at the Cu/Au^+ mass ratio of 200, final copper concentration was 0.003 mol/l higher than the initial copper concentration. In conclusion, for recycling these barren solutions at the optimum Metal/Gold mass ratio to the gold leaching stage, it could be required to make up the copper concentration for both zinc and aluminium precipitations and not required for copper precipitation.

4.5.2 Ammonia and thiosulfate concentration

Under the selected optimum condition (Section 4-5-1), the effects of Metal/Gold mass ratio on the final ammonia and

Table 2 Comparison of Three Metal Powders in Gold Cementation at the Optimum Reagent Combination of 1 mol/l NH_4OH , 0.01 mol/l $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5.

	Zn	Cu	Al
Metal/ Au^+ mass ratio (8 mg/l Au)	30	50	30
Gold recovery	100%	93%	100%
Thiosulfate consumption	Increase	Decrease	Slightly decrease
Copper concentration	Decrease to near zero	Increase greatly	Decrease slightly
Recycling barren solution	Need to replenish copper concentration	Need to replenish thiosulfate concentration	Need to replenish both copper and thiosulfate concentration

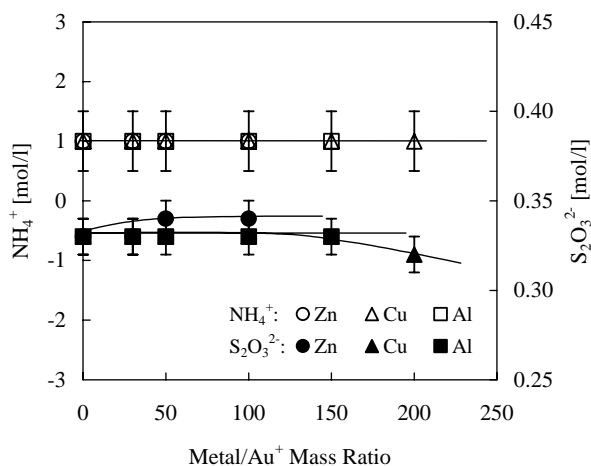
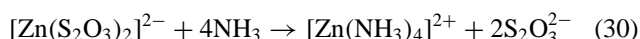


Fig. 14 Effect of Metal/ Au^+ mass ratio on thiosulfate and ammonia concentrations. 8 mg/l-Au pregnant solution with 1 mol/l NH_4OH and 0.01 mol/l CuSO_4 and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5, 60 min agitation.

thiosulfate concentrations are shown in Fig. 14.

As the Zn/Au^+ mass ratio was higher than 50, the final thiosulfate concentration increased 0.01–0.02 mol/l. This may be attributed to the formation of amine complex as shown in eqs. (13) and (30):



At copper precipitation, 0.01–0.03 mol/l $\text{S}_2\text{O}_3^{2-}$ was consumed at the Cu/Au^+ mass ratio of 200. At aluminium precipitation, the Al/Au^+ mass ratio did not significantly change the thiosulfate concentration. However, excess addition of aluminium powder to the Al/Au^+ mass ratio of 150 caused about 0.01 mol/l $\text{S}_2\text{O}_3^{2-}$ reduction. The thiosulfate consumption may be attributed to the formation of $[\text{Al}(\text{S}_2\text{O}_3)_2]^-$ as shown in eqs. (22) and (27).

Ammonia concentration maintained from the beginning to the end of gold cementation process for all three metal powders.

The comparison of three metal powders in gold cementation is shown in Table 2. The choice of metal powder depends upon the efficiency of gold recovery and the feasibility of recycling the barren solution.

5. Conclusion

Gold cementation test was carried out from an 8 mg/l Au ammonium thiosulfate solution by using 45 μm of zinc, copper and aluminium powders. The composition of the

pregnant solution were 1–5 mol/l NH_4OH , 0.01–0.05 mol/l CuSO_4 , 0.2–0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ and pH 9.5–10.5. The results demonstrated that the lower ammonia and copper concentrations and higher thiosulfate concentration could minimize the required Metal/Gold mass ratio to recover more gold. It was concluded that the optimum reagent combination for gold cementation was 1 mol/l NH_4OH , 0.01 mol/l CuSO_4 and 0.4 mol/l $(\text{NH}_4)_2\text{S}_2\text{O}_3$ at pH 9.5. Under this optimum condition, the following results were concluded.

(1) Zinc and aluminium precipitations recovered 100% of gold with Metal/Gold mass ratio of 30. The copper consumption was 0.009 mol/l by zinc precipitation and 0.007 mol/l by aluminium precipitation.

(2) Copper precipitation was the lowest copper loss with 93% of gold recovery at Cu/Au^+ mass ratio of 50. The excess addition of copper powder increased the copper concentration.

(3) At Zn/Au^+ mass ratio of 100, the thiosulfate concentration increased 0.01–0.02 mol/l at zinc precipitation. On the other hand, the thiosulfate concentration decreased 0.01–0.03 mol/l at aluminium and copper precipitations.

(4) Ammonia concentration did not changed significantly during the gold cementation process.

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