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Study on gold concentrate leaching by iodine-iodide

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Abstract: Gold extraction by iodine-iodide solution is an effective and environment-friendly method. In this study, the method using iodine-iodide for gold leaching is proved feasible through thermodynamic calculation. At the same time, experiments on flotation gold concentrates were carried out and encouraging results were obtained. Through optimizing the technological conditions, the attained high gold leaching rate is more than 85%. The optimum process conditions at 25°C are shown as follows: the initial iodine concentration is 1.0%, the iodine-to-iodide mole ratio is 1:8, the solution pH value is 7, the liquid-to-solid mass ratio is 4:1, the leaching time is 4 h, the stirring intensity is 200 r/mim, and the hydrogen peroxide consumption is 1%.

Keywords: gold extraction; leaching; iodine; iodides

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1. Introduction

Gold as a kind of unique precious metal that has played an important role since ancient times. The demand for gold is increasing with the world's economy development. Meanwhile, gold extraction technologies have been improved dramatically. There are many kinds of methods to leach gold from ores, such as cyanidation, amalgamation, thiosulfate, thiourea, lime sulfur, rhodanate, polysulfide, bromine, and iodine-iodide methods [1-4]. The most widely used method is cyanidation. Cyanidation has been invented for more than 100 years. The advantages of cyanidation include perfect process flow, higher recoveries, strong adaptability of ore, and technological simplicity [5]. The cyanidation method becomes more practiced after the further study of gold extraction from pregnant solution, such as zinc dust precipitation, carbon adsorption, ion exchange, and solvent extraction [6]. Approximately 70% of new plants are still adopting this process now [7]. However, there are several disadvantages of the cyanidation method: longer leaching time (more than 24 h; the heap leaching is even longer); higher production cost because more cyanide will be consumed when ore contains cyanicides such as copper, antimony, arsenic, iron, zinc, and sulfur [8]; and lower leaching rate for refractory gold ore. Especially, the cyanide is an extremely toxic substance and it is seriously harmful to human beings and the surrounding environment. According to the developments of science and the increasing concern of environment, these disadvantages are unbearable. New gold extraction technology is needed urgently. Compared with the cyanide, iodine is a nontoxic reagent, and it is popularly applied in medicine [7]. Gold leaching by iodine-iodide solution from the ore has been studied by some researchers worldwide [9-11], but the treatment of gold concentrate has not been researched. In this article, through the mechanism analysis and single-factor experiments, gold leaching using iodine-iodide solution was discussed on flotation gold concentrate from Shuangqishan, Fujian Province, China.

2. Theoretical basis of gold leaching by iodine-iodide solution

The stability study of gold anion complexes AuX_2 (X is an anion) by the Moscow National Precious Metal Exploration Institute showed that $CN^- >> I^- > Br^- > Cl^-$

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> NCS^- > NCO^- [12]. It meant that the strength of the gold-iodine complex is weaker than the gold-cyanide complex but stronger than bromine, chlorine, thiocyanates, and cyanates [13]. The gold dissolution chemical reactions in iodine-iodide solution are shown as follows.

$I_2(s) = I_2(l)$ (1)

$I_2(l) + I^- = I_3^-$ (2)

Anode reaction:

$Au + 2I^- = AuI_2^- + e^-$ (3)

$Au + 4I^- = AuI_4^- + 3e^-$ (4)

Cathode reaction:

$I_3^- + 2e^- = 3I^-$ (5)

Total reaction:

$2Au + I^- + I_3^- = 2AuI_2^-$ (6)

$2Au + 3I_3^- = 2AuI_4^- + I^-$ (7)

Standard Gibbs free energies of the above-mentioned species at 25°C and 0.1 MPa are shown in Table 1.

Table 1. Standard Gibbs free energy of species at 25°C and 0.1 MPa [14]

Formula	State	Δ_f $^\ominus_m$ (kJ·mol^{-1})
Au	Solid	0
I^-	Aqueous	−51 67
I_3^-	Aqueous	−51 51
AuI_2^-	Aqueous	−47 99
AuI_4^-	Aqueous	−41 00

In reaction (3), $\Delta_f G^\ominus_m = [-47.99 - 2 \times (-51.67)]$ kJ·mol^{-1} = 55.35 kJ·mol^{-1}.

Reaction (3) is a half-reaction that loses electrons; thus, $\Delta f G^\ominus_m = nF\varphi^\ominus_-$ [15],

$$\varphi^\ominus_- = \frac{\Delta_f G^\ominus_m}{nF} = \frac{55.35 \times 10^3}{1 \times 96485} \text{ V} = 0.574 \text{ V}.$$

Similarly, for reaction (4), $\varphi^\ominus_- = 0.572$ V.

In reaction (5), $\Delta_f G^\ominus_m = -103.5$ kJ·mol^{-1}.

Reaction (5) is a half-reaction that receives electronic; thus, $\Delta_f G^\ominus_m = nF\varphi^\ominus_-$ [15], $\varphi^\ominus_+ = 0.536$ V.

Because that $E^\ominus = \varphi^\ominus_+ - \varphi^\ominus_-$, $E^\ominus = -0.038$ V in reaction (6).

Similarly, $E^\ominus = -0.036$ V in reaction (7).

The Nernst equations of reactions (6) and (7) are shown as follows:

$$E = -0.038 + \frac{0.05917}{2} \lg\left\{\left[\alpha\left(I_3^-\right)\cdot\alpha\left(I^-\right)\right]/\alpha^2\left(AuI_2^-\right)\right\} \quad (8)$$

$$E = -0.036 + \frac{0.05917}{6} \lg\left\{\left[\alpha^3\left(I_3^-\right)\right]/\left[\alpha^2\left(AuI_4^-\right)\cdot\alpha\left(I^-\right)\right]\right\} \quad (9)$$

The standard electric potentials of reactions (6) and (7) are -0.038 and -0.036 V, respectively. Due to $\Delta_f G_m = -nFE$, $\Delta_f G_m > 0$, the reactions could not proceed spontaneously in the standard condition. However, their Nernst Eqs. (8) and (9) show that the concentrations of I^-, I_3^-, AuI_2^-, or AuI_4^- are changed, $\Delta_f G_m$ could become negative and the reactions could proceed spontaneously. For example, according to Eqs. (8) and (9), under the condition that $\alpha(AuI_2^-) = \alpha(AuI_4^-) = 10^{-4}$ mol·L^{-1}, $\alpha(I^-) = 0.1$ mol·L^{-1}, and $\alpha(I_3^-)$=0.1 mol·L^{-1}, the chemical potentials of reactions (6) and (7) are 0.169 and 0.013 V, respectively. The reactions could proceed spontaneously under this condition. Therefore, leaching gold from the ore using iodine-iodide solution is feasible in theory.

The electric potential-pH values (E_h-pH) diagram of the Au-I_2-I^--H_2O system also indicated that gold dissolution in iodine-iodide solution was feasible in theory. Li *et al.* [16-17] pointed out that (1) iodine-iodide solution was suitable for gold dissolution; (2) gold-iodine complex ions AuI_2^- and AuI_4^- were stable in water or hydrogen peroxide solution; (3) because of high oxidation potential, hydrogen peroxide as a auxiliary oxidant was good for gold dissolution; and (4) the pH value of iodine-iodide solution should be controlled as acid or neutral.

3. Experimental

3.1. Material source and properties

The flotation gold concentrate used in this study came from Shuangqishan, Fujian Province, China. Pyrite was the major metallic mineral; calcite and quartz were the major gangue minerals. Gold minerals were distributed nonuniformly and native gold was the main form. The dissemination size of gold was very fine (several microns). Most of the gold was filled in the cavities of pyrite in xenomorphic granular textures and filled in the fracture as short vein or granular textures. Little was distributed in quartz as xenomorphic granular textures or vein.

The chemical analysis of the gold concentrate showed that it consisted of 60.62 g/t Au, 102 g/t Ag, 0.13wt% As, 21.54wt% Fe, and 22.48wt% S.

For the mineral leaching process, when the size of ground products becomes finer, more valuable minerals are exposed, which benefits to the contact of gold and leaching solution. Therefore, the finer size of ground products could reach a higher leaching rate, but fine grinding of the ore would increase costs. Therefore, based on the leaching theory and plant actual situation, the size of ground products in this study was controlled at 99% passing 200 mesh and 80% passing 325 mesh.

3.2. Reagents and methods

Reagents used in this study included iodine, potassium iodide, hydrochloric acid, sodium hydroxide, and hydrogen peroxide (30%). All the reagents were analytically pure, and water used in the experiment was tap water in Beijing.

The leaching experiment was carried out in a 500 mL beaker, and the sample per test was 50 g. The solution was mixed with an MY3000-6 intelligent stirrer, timed with a chronograph. The pH value of the solution was adjusted by hydrochloric acid or sodium hydroxide and checked by Mettler Toledo Delta 320.

The pregnant solution was filtered after leaching and kept for extracting of gold or rebirth of iodine. The grade of gold in tailings was analyzed for calculating the leaching rate of gold.

4. Results and discussion

A series of experiments were performed under various process conditions. The process condition variables included initial iodine concentration, iodine-to-iodide mole ratio, solution pH values, liquid-to-solid mass ratio (L/S), stirring intensity, leaching time and temperature, and dosage of hydrogen peroxide. Their effects on gold leaching rate are discussed as follows.

4.1. Effect of initial iodine concentration on gold leaching rate

The gold extraction rate is different when the initial iodine concentration changes. In iodine-iodide solution, the iodine that exists in the form of I_3^- is more than 99% [10]. Based on thermodynamics analysis, the gold leaching rate is higher and higher with the increase of iodine concentration. The effect of initial iodine concentration on the extraction of gold was investigated from 0.4% to 1.4% at 25°C for 4 h at an iodine-to-iodide mole ratio of 1:8, a solution pH value of 7, a L/S ratio of 4:1, and a stirring intensity of 600 r/min. The results are shown in Fig. 1. As indicated in Fig. 1, when the initial iodine concentration changes from 0.4% to 0.8%, the gold leaching rate quickly increases at first and then slightly changes from 0.8% to 1.4%. This presents that a proper initial iodine concentration is of benefit to leaching, but too much dosage is not necessary. Consequently, the further tests were carried out by keeping the initial iodine concentration fixed at 1.0%.

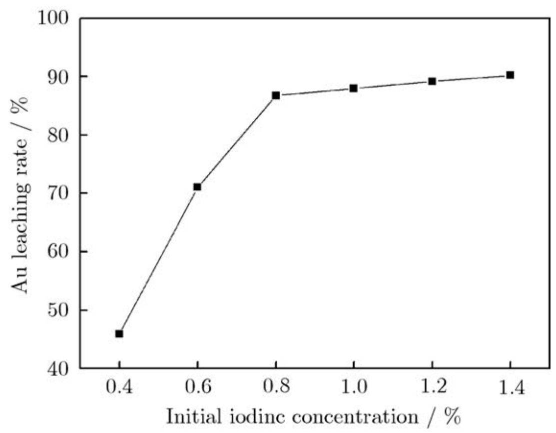

Fig. 1. Effect of initial iodine concentration on gold leaching rate.

4.2. Effect of iodine-to-iodide mole ratio on gold leaching rate

Gold can be oxidized when a complexing agent exists in the leaching solution. The oxidant and complexing agent existing together is an essential condition to gold dissolution. It is better for gold dissolution when the oxidability of the oxidant is strong and the stability constant of complexes between gold and ligands is big. During gold extraction in iodine-iodide solution, I_2 is the oxidant and I^- is the ligand [18-19]. The effect of iodine-to-iodide mole ratio on the gold leaching rate is described in Fig. 2. The iodine-to-iodide mole ratio is 1:4, 1:6, 1:8, 1:10, and 1:12, respectively. Other experiment parameters were as follows: the initial iodine concentration was 1%, the solution pH value was 7, the L/S ratio was 4:1, the leaching time was 4 h, the stirring intensity was 600 r/min, and the temperature was 25°C. As shown in Fig. 2, the gold leaching rate increases as the iodine-to-iodide mole ratio decreases. Hence, the iodine-to-iodide mole ratio should be as low as possible in practice. However, the gold leaching rate has no obvious change when the iodine-to-iodide mole ratio was lower than 1:8. Considering the costs, all further experiments were performed at the iodine-to-iodide mole ratio of 1:8.

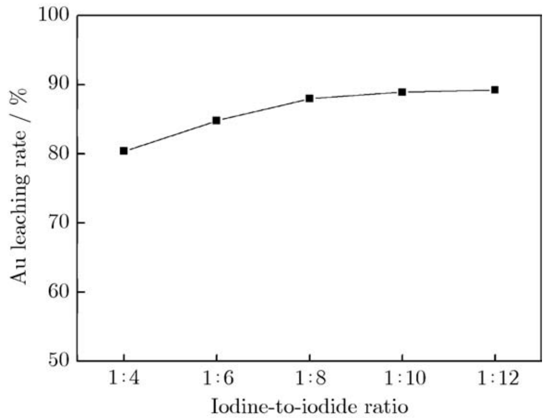

Fig. 2. Effect of iodine-to-iodide mole ratio on gold leaching rate.

4.3. Effect of pH value on gold leaching rate

The adjusting agents of pH values in this study were hydrochloric acid (1%) and sodium hydroxide (10%). The experiment conditions were shown as follows: the pH value was varied from 4 to 10 (the interval was 1.5), the initial iodine concentration was 1%, the iodine-to-iodide mole ratio was 1:8, the L/S ratio was 4:1, the leaching time was 4 h, the stirring intensity was 600 r/min, and the temperature was 25°C. The experimental results are shown in Fig. 3. As presented in Fig. 3, the leaching rate of gold keeps at a high level and increases before the solution pH value reaches 8.5. However, it decreases quickly when the pH value is higher than 8.5. In the Au-I_2-I^- system, gold dissolution is the coefficient result of I_3^- and I^-, which are both irreplaceable. However, when the pH value is in a relatively high level, the I_3^- concentration decreases and the gold dissolution rate also decreases; thus, the gold leaching rate will reduce [20]. Therefore, the proper condition of gold leaching in iodine-iodide solution is neutral or acidic, but considering the equipment corrosion and safety production of a plant, the value of 7 is adopted as the solution pH value.

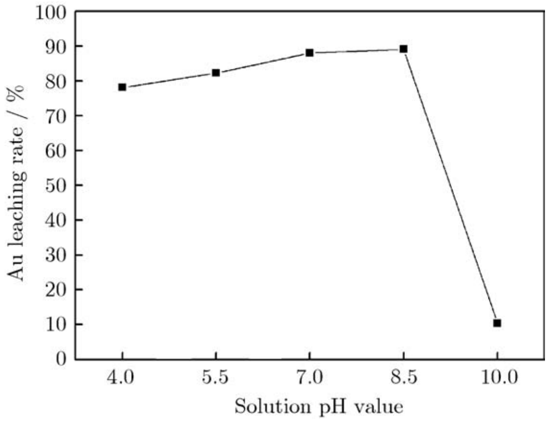

Fig. 3. Effect of solution pH values on gold leaching rate.

4.4. Effect of L/S ratio on gold leaching rate

To study the effect of L/S ratio on the gold extraction, the L/S ratio was varied from 2:1 to 6:1. Other variables were as follows: the initial iodine concentration was 1%, the iodine-iodide ratio was 1:8, the solution pH value was 7, the leaching time was 4 h, the stirring intensity was 600 r/min, and the temperature was 25°C. The experimental results are shown in Fig. 4. It can be observed that the gold leaching rate gradually increases with the increase of L/S ratio. The reason is that a lower pulp density means a lower pulp viscosity, and in this condition, diffusion velocity from the reagent to the surface of gold becomes much faster, thus the dissolution speed of gold increases, then the extraction rate of gold increases. However, a higher L/S ratio means a more reagent consumption, a more investment of equipment, and a more difficulty of the recovery of iodine. Considering these factors, 4:1 is used as the L/S ratio.

4.5. Effect of leaching time on gold leaching rate

The effect of leaching time on the gold leaching rate is shown in Fig. 5. The leaching time was 2, 4, 6, 8, and 10 h, respectively. Other experimental parameters were as follows: the initial iodine concentration was 1%, the iodine-to-iodide mole ratio was 1:8, the L/S ratio was 4:1, the solution pH value was 7, the stirring intensity was 600 r/min, and the temperature was 25°C. As shown in Fig. 5, the gold leaching rate is higher and higher with the increase of leaching time. When the reaction time reaches 4 h, the leaching rate reaches up to a relatively high level. However, when the time is varied from 4 to 10 h, the leaching rate has almost no change. In this case, 4 h is selected as the optimum value in the following study.

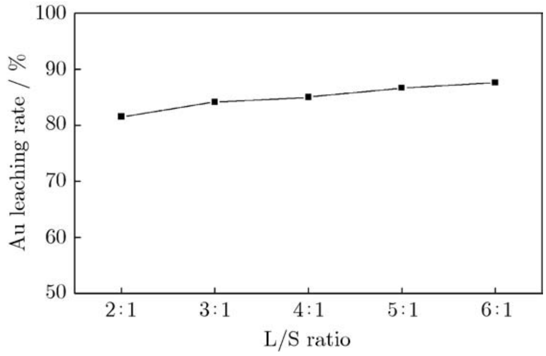

Fig. 4. Effect of L/S ratio on gold leaching rate.

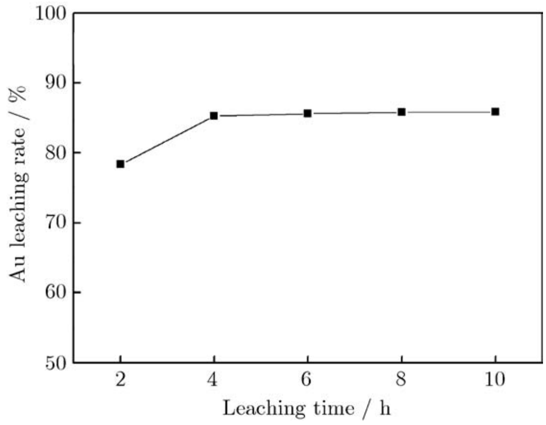

Fig. 5. Effect of leaching time on gold leaching rate.

4.6. Effect of stirring intensity ratio on gold leaching rate

The main purpose of stirring is to increase the contact frequency between the ore and leaching solution to enhance the leaching rate. Maintaining the initial iodine concentration of 1%, the iodine-to-iodide mole ratio of 1:8, the L/S ratio of 4:1, the solution pH value of 7, the leaching time of 4 h, and the temperature of 25°C, the stirring intensity was varied from 200 to 800 r/min. The results shown in Fig. 6 indicate that gold leaching rate gradually decreases with the increase of stirring speed. It is possible that the ore and liquid will rotate together in the container when the mixing speed is higher. Then, the ore and liquid could not mix uniformly, and the leaching solution on the ore surface could not be updated. Therefore, the reaction proceeding will be influenced [21]. When the stirring speed is 200 r/min, the leaching rate is the highest. However, when the stirring speed was less than 200 r/min, there was sediment that appeared in the ore pulp based on the experiment phenomenon. Therefore, 200 r/min was adopted as the stirring intensity in the further study.

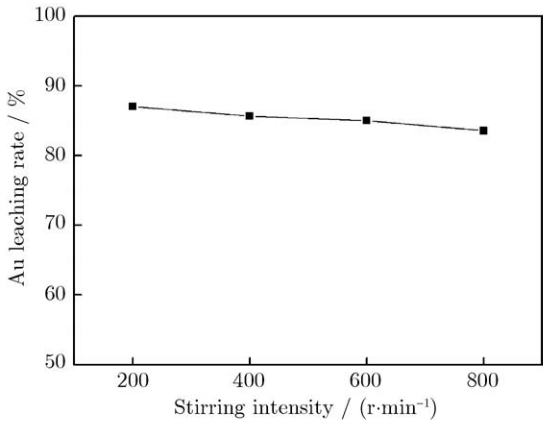

Fig. 6. Effect of stirring intensity on gold leaching rate.

4.7. Effect of leaching temperature on gold leaching rate

Temperature has an important impact on the chemical reaction and the diffusion rate for most leaching processes. When the energy stored in the mineral grain increases, the power of destroying the mineral chemical bond enhances, and the number of molecules whose kinetic energy greater than activation energy increases; therefore, the leaching process is accelerated [21]. Fig. 7 shows the effect of temperature on the gold leaching rate. Other experimental parameters were as follows: the initial iodine concentration was 1%, the iodine-to-iodide mole ratio was 1:8, the solution pH value was 7, the L/S ratio was 4:1, the leaching time was 4 h, and the stirring intensity was 200 r/min. As presented in Fig. 7, the gold leaching rate increases as the temperature increases from 15°C to 45°C. When the temperature reaches 25°C, the leaching rate reaches up to a relatively high level. However, the leaching rate changes slowly from 25°C to 45°C. Meanwhile, in view of the convenience of the experiment process, 25°C is chosen for the leaching temperature.

4.8. Effect of hydrogen peroxide consumption on gold leaching rate

Because the leaching reaction can be accelerated by adding an auxiliary oxidant in the leaching solution, hydrogen peroxide was selected as an auxiliary oxidant in this study. The effect of hydrogen peroxide concentration on the gold leaching rate was examined from 0 to 4%, whereas the initial iodine concentration, iodine-to-iodide mole ratio, solution pH value, L/S ratio, leaching time, stirring intensity, and leaching temperature were kept constant as 1%, 1:8, 7, 4:1, 4 h, 200 r/min, and 25°C, respectively. As shown in Fig. 8, when the hydrogen peroxide consumption is 1%, the leaching rate is the highest. It indicates that a proper concentration of hydrogen peroxide can accelerate the dissolution of gold. However, if it is too much, such as 2%, the leaching rate starts to decrease. The reason is that the ions of I^- and I_3^- in the solution will be oxidized by the excessive hydrogen peroxide. Thus, the leaching reaction will be destroyed; the gold leaching rate will decrease. Therefore, 1% of the hydrogen peroxide consumption is adopted in the experiments.

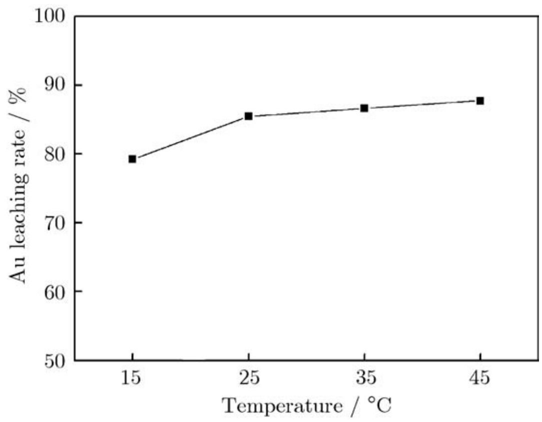

Fig. 7. Effect of leaching temperature on gold leaching rate.

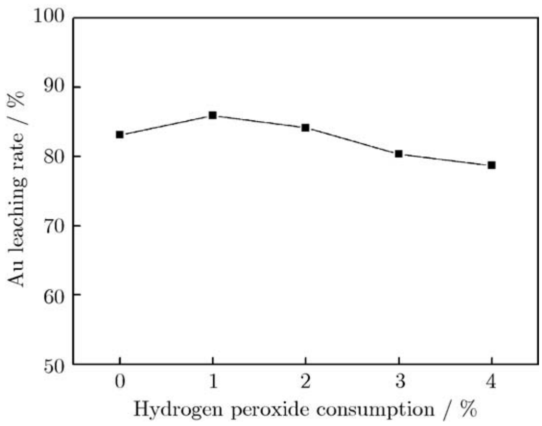

Fig. 8. Effect of hydrogen peroxide consumption on gold leaching rate.

5. Conclusions

(1) Thermodynamic calculations show that the gold leaching reactions could not proceed spontaneously in the standard condition. However, if the concentration of ions changes appropriately, the reactions can proceed spontaneously. Therefore, gold leaching from the ore in iodine-iodide solution is feasible in theory.

(2) Experimental results prove that the theory of gold leaching using iodine-iodide solution is also suitable for

treating the practical flotation gold concentrate. Through optimizing the process conditions, the gold extraction could get up to 85%, and the optimum conditions are shown as follows: the initial iodine concentration is 1.0%, the iodine-to-iodide mole ratio is 1:8, the solution pH value is 7, the L/S ratio is 4:1, the leaching time is 4 h, the stirring intensity is 200 r/min, the leaching temperature is 25°C, and the hydrogen peroxide consumption is 1%.

(3) The leaching rate of gold in iodine-iodide solution is much faster than in cyanidation solution. Also, iodine-iodide solution is environment-friendly.

(4) In practice, the cost of iodine and iodide might be expensive, but recycling could reduce the cost. In the future, further investigations will be done on the recovery of iodine from the wastewater and the influence of recycling iodine-iodide solution on gold leaching.

Acknowledgements

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References

[1] J.Y. Lv and G.C. Li, Research progress of several typical non-cyanide gold leaching technologies, *Mod. Min.*, 25(2009), No. 8, p. 11.

[2] G.C. Li and S.C. Lu, The development of non-cyanidation gold lixiviating technology, *China Min. Mag.*, 12(2003), No. 3, p. 1.

[3] H. Yu and X.Z. Hu, Review of gold-leaching technologies by cyanidation and non-cyanidation, *Yunnan. Metall.*, 39(2010), No. 3, p. 9.

[4] C.L. Jian, Review on extracting methods of gold, *Hydrometall. China,* 27(2008), No. 1, p. 1.

[5] J.Y. Lv, *Study on the Mechanism and Technical Conditions of Iodine Leaching of Gold* [Dissertation], Heilongjiang Institute of Science and Technology, Harbin, 2010, p. 2.

[6] Q. Xu, *Study on Gold Leaching from Waste Printed Circuit Board by Iodine Process* [Dissertation], Donghua University, Shanghai, 2009, p. 11.

[7] G.C. Li and S.C. Lu, Research on iodine leaching of gold, *Min. Metall. Eng.*, 23(2003), No. 6, p. 53.

[8] X. Dai and M.I. Jeffrey, The effect of sulfide minerals on the leaching of gold in aerated cyanide solutions, *Hydrometallurgy*, 82(2006), No. 3-4, p. 118.

[9] P.H. Qi and J.B. Hiskey, Dissolution kinetics of gold in iodide solutions, *Hydrometallurgy*, 27(1991), No. 1, p. 47.

[10] T.N. Angelidis, K.A. Kydros, and K.A. Matis, A fundamental rotating disk study of gold dissolution in iodine-iodide solutions, *Hydrometallurgy*, 34(1993), No. 1, p. 49.

[11] P.H. Qi and J.B. Hiskey, Electrochemical behavior of gold in iodide solutions, *Hydrometallurgy*, 32(1993), No. 2, p. 161.

[12] G.V. Sedelnikova and G.S. Krylov, Iodinated and brominated solvent of gold, *Schnigri*, 3(2001), p. 43.

[13] G.C. Li and S.C. Lu, Iodine leaching of gold from lean gold ore with charcoal, *J. Univ. Sci. Technol. Being*, 25(2003), No. 6, p. 501.

[14] T. Jiang, *Chemistry of Gold Extraction,* Hunan Science and Technology Press, Changsha, 1991, p. 330.

[15] Y.L. Zhang, G. Fan, and J.J. Sun, Explore formula of calculated electrode potential with thermodynamics method, *Guangzhou Chem. Ind.*, 39(2011), No.16, p.180.

[16] G.C. Li and S.C. Lu, Iodide leaching gold from carbonaceous refractory gold ores, [in] *Proceedings of the 24th International Mineral Processing Conference*, Beijing, 2008, p. 3010.

[17] G.C. Li and J.Y. Lv, Basic theory of iodide leaching of gold process, *J. Heilongjiang Inst. Sci. Technol.*, 19(2009), No. 5, p. 345.

[18] A. Davis, T. Tran, and D.R. Young, Solution chemistry of iodide leaching of gold, *Hydrometallurgy*, 32(1993), No. 2, p. 143.

[19] Z.Y. Fang, *Leaching*, Metallurgical Industry Press, Beijing, 2007, p. 20.

[20] G.C. Li, *Study on the Iodide-Iodine Leaching Technology and Theory of Unmanageable Gold Ore Containing Carbon* [Dissertation], University of Science and Technology Beijing, Beijing, 2003, p. 81-85.

[21] The Compiled Committee of Leaching Mineral Technology, *The Technology of Leaching Mineral*, Atomic energy Press, Beijing, 1994, p. 114.