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ADSORPTION-DESORPTION OF GOLD ONTO ACTIVATED CARBON FROM CYANIDE-GLYCINE LEACH SOLUTION OF COMPLEX GOLD ORE

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ABSTRACT

This research investigated the adsorption of gold on activated carbon from a cyanide-glycine leach solution and their desorption. The pH value varied between 9 and 11, and at each pH value, the concentration of activated carbon was changed between 5 and 20 g L^{-1}. The results showed that both pH and carbon concentration did not have a significant effect on gold adsorption. In all experiments, a high adsorption value of gold (above 98 %) was obtained. In all carbon concentrations, except for 5 g L^{-1}, the maximum gold adsorption was obtained at a pH of 10.5. The reduction of gold adsorption on activated carbon with the increase of pH to 11 can be related to the decrease of active points on the carbon surface due to the increase in the concentration of hydroxide ions. When the concentration of activated carbon was 5 g L^{-1}, more than 98 % of gold was adsorbed. So, a further increase in carbon concentration is not required. Elution experiments were carried out after different leaching methods; carbon in pulp (CIP) and carbon in leach (CIL). The results showed that for both leaching methods, most of the gold was dissolved in the first 6 h of the elution process and the final gold desorption for CIP and CIL were 82.89 % and 72.64 % respectively.

Keywords: gold ore, leaching, activated carbon, glycine, cyanide.

INTRODUCTION

Adsorption on activated carbon is a current method of gold recovery from alkaline cyanide leach solutions due to its simplicity, cheapness and selectivity [1]. Activated carbon is a highly porous carbonaceous material with millions of tiny interlocking pores [2]. Active carbons have micropores (< 2 nm), mesopores (2 - 50 nm), and macropores (> 50 nm) in their tridisperse microcrystalline structure [3]. Macropores on the outside of active carbon particles are a pathway for adsorption molecules, which can be pulled to pores at the core of active carbon. The surface area of most commercial activated carbon adsorbents is 800 to 1500 m^2 g^{-1}. The ability of activated carbon to adsorb metals is due to the inherent nature of ion exchange characteristics resulting from the presence of heteroatoms bonded in the form of carbon-oxygen, carbon-nitrogen, and carbon-sulfur functional groups on the carbon surface. As oxygen has a significant influence on the adsorption capacity of active carbon, the carbon-oxygen groups are the most important [4]. The acidic carbon-oxygen surface group makes the carbon surface hydrophilic and increases the adsorption of metal actions [5].

It is not well understood how the carbon surface interacts with the gold cyanide complex. Various researchers have reported several mechanisms. The four categories can summarize all the proposed theories to date. (i) the reduction theory with carbon as the reducing agent, (ii) formation and precipitation of carbonyl aurocyanide compound $[AuCN.CO(CN)_2]$ on the carbon surface, (iii) adsorption of gold as the aurocyanide complex $[Au(CN)_2]^-$ and (iv) adsorption of the gold complex with other metallic ions [6].

Recently, research has been done to use glycine as a substitute for cyanide. The results have shown that the simultaneous use of cyanide and glycine increases the dissolution rate of gold [7 - 9].

Recent research has shown that activated carbon can extract the gold glycinate complex in the presence and absence of copper from alkaline solutions [7, 10, 11]. The activated carbon adsorption technique can be used for gold recovery from glycine cyanide leach solutions, and no changes to the downstream processing are required.

The leaching of gold, silver, and their alloys in alkaline glycine-peroxide solutions and their adsorption on carbon was investigated. The determination of the carbon activity constant using the Fleming k, n method was used to evaluate the kinetics of adsorption of gold and silver to activated carbon from glycine solutions. The results showed that, similar to cyanide systems, the carbon adsorbed gold (k = 460) significantly faster than silver (k = 110) [7].

The adsorption mechanism of a gold glycine complex into activated carbon in pure alkaline glycine/glycinate systems under various process conditions and in the presence of copper was studied. The results showed that the adsorption capacity increased with increasing free glycine, copper and calcium chloride concentration but decreased with increasing solution pH and initial gold concentration [10]. The carbon load increases significantly with an increase in glycine concentration up to 10 g L^{-1}. Increasing pH from 10 to 11 to 12 decreased the gold adsorption, as more hydrogen ions will compete with gold ion complexes for active sites on the carbon. The gold adsorption was reduced by more than 50 % with an increase in initial gold concentration from 2 to 16 ppm. Surprisingly, with increasing copper concentrations in the solution, the gold loading capacity of the carbon was increased. Instead of competing with gold complexes for adsorption on activated carbon, copper complexes increase the adsorption loading of gold complexes. The adsorption of gold glycinate complexes on carbon was raised by a higher ionic strength [10].

The kinetics of gold and copper adsorption from synthetic alkaline glycine solutions onto activated carbon were determined and compared. The effect of pH, free glycine, ionic strength, initial gold, copper concentrations, and carbon concentration on the adsorption kinetics were investigated. The results showed that more than 98 % of gold was adsorbed on activated carbon in less than 6 h. The carbon concentration had a significant impact on the initial gold adsorption rate. Under optimal conditions, in a solution containing 2 ppm gold and 5 ppm copper, the adsorption values of gold and copper on carbon were 99 % and 15 %, respectively, which indicates the high selectivity of carbon for the gold glycinate complexes [11].

The adsorption efficiency of gold and copper onto activated carbon from cyanide-starved alkaline glycine solutions in terms of equilibrium loading isotherms and kinetics was studied. Results showed in the presence of copper, the equilibrium gold loading increased with increasing glycine and calcium ion concentrations, and decreased with increasing cyanide concentration and did not depend on initial gold concentration [5]. The effects of glycine concentration, solution pH, cyanide concentration, initial gold concentration, adsorbent concentration, and ionic strength of the solution on the adsorption of gold and copper on activated carbon were studied. The results have shown that among these parameters, only carbon concentration had a significant effect on gold adsorption. With the increase of carbon concentration from 2 g L^{-1} to 4 g L^{-1}, the gold adsorption on activated carbon increased from 91.8 % to 97.9 %. Gold yields of 98 % or above have been achieved by changing all other parameters. The adsorption tendency towards gold as opposed to copper was also observed for active carbon. Although the copper concentration in the solution was more than one hundred times the gold concentration of gold (2 ppm gold, 300 ppm copper and no free cyanide), it recovered on average only 50 % [12].

To the best of the author's knowledge, previous research has been conducted on synthetic alkaline glycine solutions [7, 10, 11] or alkaline cyanide-glycine solutions [5, 12] and none of the research has investigated the adsorption-desorption on gold onto activated carbon from cyanide-glycine leach solution of gold ore. In this research a complex gold ore was leached using a synergistic lixiviant mixture of glycine and cyanide and activated carbon was used as a potential adsorbent agent for the recovery of gold from the leach solution. The effects of pH and carbon concentration on the adsorption process, and the subsequent elution of the gold complexes on granular activated carbon, were assessed.

EXPERIMENTAL

Materials

Samples were taken from the Gandi gold mine in Semnan Province, Iran, where gold appeared as free metal, part of sulfide minerals, and as petzite (Te, Ag, Au) [13]. 15 kg samples from various mine areas were combined, ground to pass 74 μm, and subsampled to

214 g using riffle sampling. The ore was purified with a Mozley table, and X-ray diffraction (XRD) was used to identify minerals, revealing chalcopyrite, quartz, calcite, kaolinite, and galena. Elemental analysis was performed via acid digestion and ICP-OES, while gold content was determined by fire assay and Atomic Absorption Spectroscopy (AAS). The sample contained 5.2 ppm Au, 5.3 ppm Ag, 1039 ppm Cu, 3910 ppm Zn, and 10667 ppm Pb.

Leaching and adsorption experiments

All leaching experiments were carried out under the optimal conditions obtained from the previous research [8]. In all leaching experiments, subsamples weighing 214 g and 500 cc deionized water (30 % solid weight) were poured into 2.5 L Winchester bottles. The pH of the pulp was adjusted to 11 by adding NaOH. Then, 0.01 mol L^{-1} (1145 ppm) sodium cyanide, 0.06 mol L^{-1} (10521 ppm) glycine, and 2 % hydrogen peroxide (H_2O_2) were added to the contents of the bottle. All reagents used in this research were analytical grade and manufactured by Sigma-Aldrich. The pulp was agitated by rolling the bottle on a roller at room temperature and a rotation speed of 150 rpm (Fig. 1). After 24 h the pulp was filtered, and the dried cake and solution were analysed for gold and copper using atomic adsorption spectrometry (AAS).

In adsorption experiments, which are a major part of this research, first, the pregnant solution obtained from the leaching step was poured into a 2.5 L Winchester bottle (Fig. 1). Then, the pH of the solution was adjusted to the desired value. Next, some activated carbon (coconut based, Jacobi, 6 - 12 mesh) was poured to the bottle content. The activated carbon was already washed with distilled water and then dried in an oven at 70°C and cooled at ambient temperature in a desiccator. The activated carbon suspension was agitated by rolling the bottle on a roller at room temperature, and a rotation speed of 150 rpm. After 24 h, the solution containing activated carbon was filtered and a five cc representative sample of the solution was sent to the laboratory for gold and copper analysis. The percentage of metal adsorbed on activated carbon was calculated by the following equation:

$$Metal\ adsorbed\ on\ carbon\ \% = 100 - \\ - \frac{metal\ content\ in\ solution\ after\ carbon\ absorption}{metal\ content\ in\ solution\ before\ carbon\ absorption} \times 100 \tag{1}$$

Fig. 1. Batch leaching and adsorption experiments setup.

Table 1. Elemental content of the mineral sample.

Concentration, ppm	Element	Concentration, ppm	Element	Concentration, ppm	Element
7793	S	28193	Fe	5.3	Ag
23.2	Sb	9	La	33.14	Al
5.6	Sc	5	Li	256	As
19.1	Th	1547	Mg	34375	Ca
33	V	1488	Mn	10.5	Cd
8	Y	6.1	Mo	18	Ce
0.5	Yb	57	Ni	20	Co
3910	Zn	585	P	19	Cr
5.2	Au	10667	Pb	1039	Cu

Elution experiments

Elution is a process where the gold on the activated carbon particles is desorbed into an aqueous solution. Two industrial elution methods are the Zadra process and the Anglo American Research Laboratories (AARL) process [14].

In the Zadra process, the gold cyanide-loaded activated carbon is stripped with a hot solution (85 - 95°C) of 1 - 2 % NaOH and 0.2 - 0.5 % NaCN at atmospheric pressure. The process has a low capital cost, and elution duration is typically 18 hours [15].

The AARL process starts with a dilute hydrochloric acid wash followed by a water wash to remove residual acid. Then, the carbon is washed with a solution containing about 3 % sodium cyanide and 1 % sodium hydroxide for about 30 min at a temperature of 110°C. Next, the carbon is eluted with high quality softened water at a temperature of 120°C. Finally, gold is recovered from the pregnant eluant by electrowinning and the barren eluant is discarded [16]. In AARL, electrowinning process is done separately after the elution process, while in the Zadra process, elution and electrowinning is done simultaneously [17].The advantages of the AARL are its relative speed and the decoupling from electrowinning. The AARL requires less heat input. The advantage of the Zadra is a simpler flowsheet [18].

Elution experiments were carried out after different leaching methods; carbon in pulp (CIP) and carbon in leach (CIL). In both situations, the carbon content was 5 g L^{-1}, the elution time was 24 h, and the pH of the gold adsorption on carbon for the CIP condition was 11.

Batch elution experiments were performed in a round-bottomed magnetically stirred flask with a reflux condenser (Fig. 2). The flask was placed in a paraffin oil bath, and then the bath was put on a heater stirrer to agitate the content and increase the reaction temperature. The temperature of the oil bath was controlled by a mercury thermometer. The reflux of the reactor was cooled by water recirculation around the condenser. The fresh eluant was made up using 2 % NaOH and 0.5 % NaCN solution. Typically, 5 g of loaded carbon was contacted with 100 cc of eluent solution for 24 h. During the elution experiment, the solution was sampled at several intervals until 24 h and analysed for gold by atomic adsorption spectrophotometry. The percentage of gold desorption from activated (gold elution %)) carbon was calculated by the following equation:

$$\text{Gold elution from carbon } \% = \frac{\text{metal content in solution after elution}}{\text{metal content adsorbed on carbon}} \times 100 \qquad (2)$$

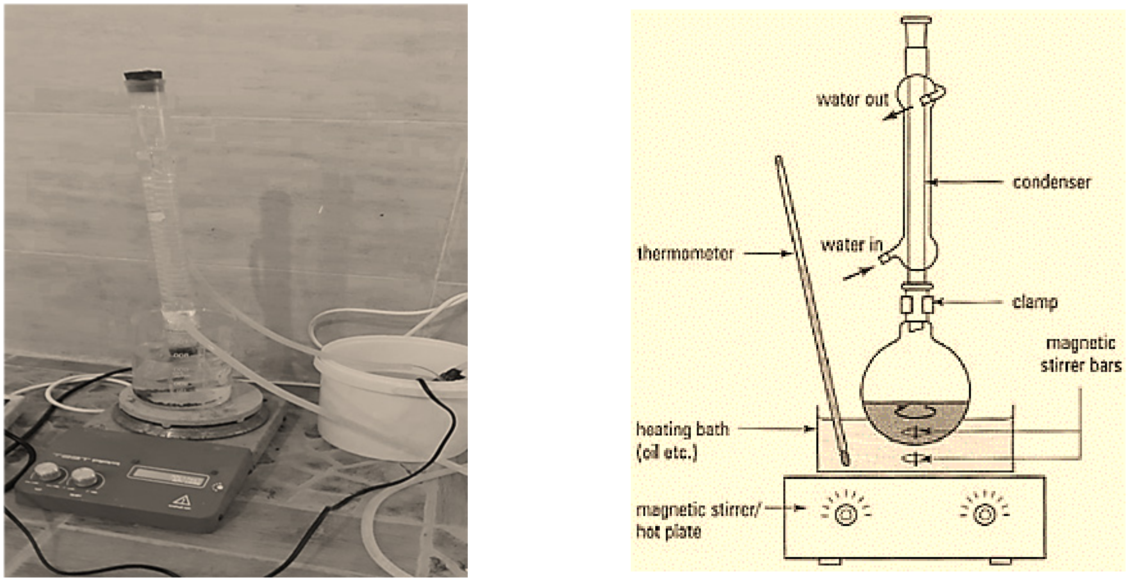

Fig. 2. Batch elution experiments setup.

RESULTS AND DISCUSSION

Leaching and adsorption experiments

Table 2 shows the plan of leaching and carbon adsorption experiments. It can be seen that all leaching experiments were carried out under the optimal conditions obtained from the previous research [8]. However, in this research, the leaching time was reduced to 24 to reduce the time of the experiments. The leaching experiment results show that the gold dissolution is about 31.4 %.

In the carbon adsorption experiments, two parameters, pH and carbon content, were changed, and their effects were investigated. The pH value was varied between 9 and 11. At each pH value, the amount of activated carbon was changed between 5 and 20 g L^{-1}. Other conditions were the same in all carbon adsorption experiments.

The carbon adsorption results (Table 2) show that pH and carbon content parameters had no significant effect on the percentage of carbon adsorption. In all adsorption experiments, a high amount of gold adsorption (above 98 %) was obtained. The minimum gold adsorption value is 98.04 % (experiment No. 15) and the maximum adsorption value is 99.77 % (experiment No. 12). The most significant difference between the results is only 1.73 %, which is very low. However, the effect of two parameters, pH and carbon content, are discussed as follows.

Various researches have shown that the pH of the solution is one of the critical parameters affecting the adsorption of gold on activated carbon [19, 20]. Changes in the pH value also affect the formation of different glycine complexes. Glycine is formed in three forms: cation ($^+H_3NCH_2COOH$), anion ($H_2NCH_2COO^-$), and zwitterion ($^+H_3NCH_2COO^-$) in solution. The cationic form is often created at pHs below 2.35, the anionic is often formed at pHs above 9.78, and the neutral form is often created at pHs between 2.35 and 9.78 [21, 22]. Fig. 3 shows the effect of pH changes in different amounts of carbon on gold adsorption. It can be seen that, except for the carbon content of 5 g L^{-1}, in other conditions, the highest amount of gold adsorption was obtained at a pH of 10.5. However, the percentage of gold adsorption is above 98 % at all pHs. In similar research, the adsorption of gold complexes from glycine leach solution on activated carbon was investigated. The results showed that at pH 9, 10, 11, and 12, 100 % of gold

Table 2. Plan of leaching and adsorption experiments and their results.

No	leaching step					adsorption step		
	Glycine, mol L^{-1}	Cyanide, mol L^{-1}	H_2O_2, %	pH	Au dissolution, %	pH	Carbon, g L^{-1}	Au adsorption, %
1	0.06	0.01	2	11	30.98	9	5	99.48
2	0.06	0.01	2	11	31.44	9	10	99.38
3	0.06	0.01	2	11	32.11	9	15	98.55
4	0.06	0.01	2	11	29.24	9	20	98.90
5	0.06	0.01	2	11	33.23	10	5	98.74
6	0.06	0.01	2	11	32.40	10	10	99.05
7	0.06	0.01	2	11	28.74	10	15	99.32
8	0.06	0.01	2	11	30.08	10	20	99.44
9	0.06	0.01	2	11	32.33	10.5	5	98.63
10	0.06	0.01	2	11	33.35	10.5	10	99.51
11	0.06	0.01	2	11	32.11	10.5	15	99.36
12	0.06	0.01	2	11	31.38	10.5	20	99.77
13	0.06	0.01	2	11	29.24	11	5	99.20
14	0.06	0.01	2	11	32.06	11	10	98.99
15	0.06	0.01	2	11	31.95	11	15	98.04
16	0.06	0.01	2	11	31.86	11	20	98.41

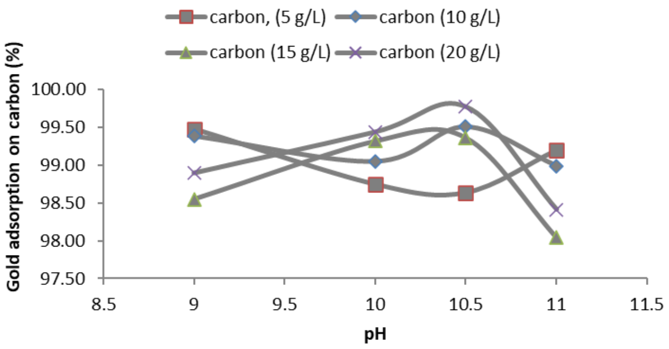

Fig. 4. Effect of carbon content on gold adsorption on activated carbon.

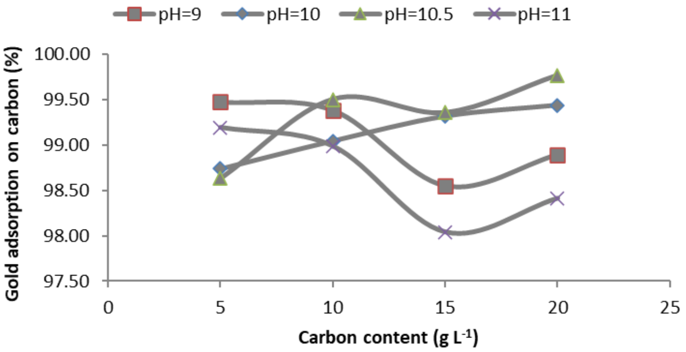

Fig. 3. Effect of pH on gold adsorption on activated carbon.

was adsorbed after 6 h. However, the highest adsorption rate was obtained at pH 10 [5]. The reduction of gold adsorption on activated carbon with increasing pH to 11 can be related to the decrease of active points on the carbon surface due to the increase in the concentration of hydroxide ions [23]. At very high pHs, hydroxide ions increase and compete with gold complexes to sit on the activated carbon surfaces [24].

Fig. 4 shows the effect of activated carbon content on gold adsorption. It can be seen that at pH 10 and 10.5, with the increase in the content of carbon, the gold adsorption has an almost upward trend, but at pH 9 and 11, with the increase in the content of carbon from 5 to 15 g L^{-1}, the adsorption of gold decreases and then It has increased by increasing the carbon content from 15 to 20 g L^{-1}. It can be concluded that 5 g L^{-1} of carbon is enough to adsorb gold, and gold adsorption of more than 98 % is obtained, and there is no need to increase carbon. Also, the possibility of further reducing the carbon content can be investigated.

Elution experiments

Fig. 5 shows the gold desorption from activated carbon (elution) for CIP and CIL experiments. In both experiments, most of the gold is dissolved in the first 6 h, with the increase of elution time to 24 h, the gold desorption increases by only 10 %. The final gold desorption for CIP (82.89 %) is about 10 % more than CIL (72.64 %). This difference may go back to the adsorption step. In CIL, when the slurry first meets activated carbon gold has not yet sufficiently leached, so there is less competition between gold complexes for adsorption on carbon, and gold complexes have a better chance to be pulled to pores at the core of active

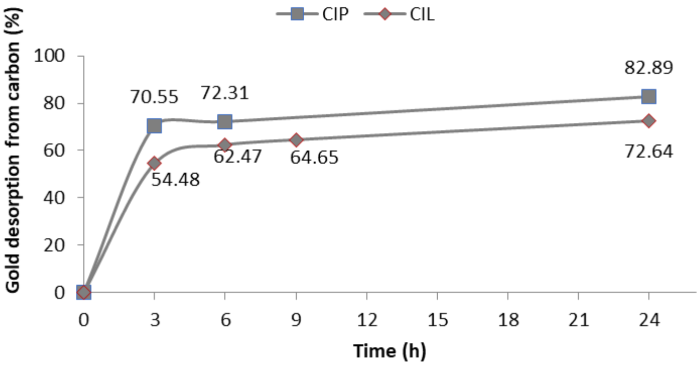

Fig. 5. Gold desorption from activated carbon.

carbon; therefore, it is more difficult to separate them from carbon pores in the elution process.

CONCLUSIONS

In all experiments, a high adsorption value of gold (above 98 %) was obtained. Except for the carbon content of 5 g L^{-1}, in other conditions, the highest amount of gold adsorption was obtained at a pH of 10.5. The reduction of gold adsorption on activated carbon with increasing pH to 11 can be related to the decrease of active points on the carbon surface due to the increase in the concentration of hydroxide ions. The gold desorption for CIP and CIL were 82.89 % and 72.64 % respectively. Most of the gold was dissolved in the first 6 hours of the elution process. It can be concluded that the gold elution process from carbon is successful for the cyanide-glycine leaching of gold ore. However, further research should be done on different iron ores to clarify different aspects of the process.

Authors' contributions: *F.F.: conducted the majority of the experimental tests and performed data analysis. K.B.: conceptualized the research idea and contributed to both data analysis and manuscript preparation. R.G.: provided the necessary resources and facilitated the interpretation of the results. All authors contributed to the discussion of the findings, reviewed the manuscript, and approved the final version.*

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