

Article

A Study on the Combined Flotation–Roasting–Leaching Process for Treating High-Carbon Gold Ore and Its Kinetics

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Abstract

Direct cyanide leaching of high-carbon gold ores typically results in poor gold extraction due to the “preg-robbing” effect of carbonaceous matter. This study investigates a refractory high-carbon gold ore from Laos. Based on detailed process mineralogy that identified the occurrence state of gold and the distribution of harmful impurities, a combined process consisting of flotation pre-concentration, oxidative roasting, and a Mill–Leach Coordination (MCL) process was developed. The MCL process, which integrates simultaneous grinding and leaching, continuously renews the particle surface and eliminates product-layer diffusion resistance, thereby significantly enhancing gold dissolution efficiency. Kinetic studies demonstrate that the leaching reaction follows the Avrami model and is controlled by internal diffusion, with a derived semi-empirical kinetic equation. This research provides a novel and efficient technical route for treating high-carbon gold ores and offers theoretical guidance for industrial scale-up.

Keywords: refractory gold ore; roasting; leaching kinetics; Mill–Leach Coordination (MCL)

1. Introduction

Gold is a global strategic resource possessing both commodity and financial attributes. As a commodity, gold has long been regarded as a symbol of social status and wealth due to its scarcity. Moreover, owing to its excellent chemical stability and ductility, it is widely used in electronics, aerospace, medical and health fields, among others [1–3]. In recent years, amid global geopolitical instability, the price of gold has surged, further highlighting its significance.

The high demand for gold resources has driven the exploitation and utilization of gold deposits, consuming large amounts of easily extractable gold resources. As a result, refractory gold ores have increasingly become an indispensable source of gold. Refractory gold ores account for approximately two-thirds of the world's total gold reserves [4]. Based on their mineralogical characteristics, refractory gold ores can be classified into poly-metallic sulfide ores, carbonaceous gold ores, telluride ores, etc. [5,6]. Among these, carbonaceous gold ores refer to gold ores with a high content of carbonaceous matter, which are often associated with minerals such as quartz, silicates, sulfides, and carbonates. The carbonaceous material in these ores exhibits strong adsorption capacity. During the cyanide leaching process, it adsorbs gold from the solution, causing dissolved gold to return to the leaching residue, thereby reducing the gold leaching efficiency. This phenomenon is



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known as “preg-robbing” [7–9]. Studies have shown that when the organic carbon content in the ore exceeds 0.2%, the gold leaching rate decreases significantly. In addition, sulfides such as pyrite, sphalerite, and chalcopyrite in the ore consume oxygen and cyanide during cyanide leaching due to oxidation, further inhibiting gold dissolution [10,11].

To address the issue of low gold leaching efficiency in refractory carbonaceous gold ores, pretreatment is generally required prior to cyanide leaching. This involves the use of mechanical, physical, and chemical methods to disrupt the gold-bearing minerals, thereby liberating the encapsulated gold, while simultaneously eliminating the adverse effects of harmful impurities such as carbonaceous matter on gold leaching, thus improving the leaching rate. Common pretreatment methods include: roasting, chemical oxidation, bio-oxidation, covering and inhibition, flotation, etc. [12–14]. Roasting oxidation is one of the most mature, technologically reliable, and widely applicable methods for treating carbonaceous gold ores. Under high-temperature oxidizing conditions, the organic carbon, elemental carbon, and sulfur in sulfides within the ore are oxidized and removed as gases, eliminating the detrimental effects of carbonaceous matter and sulfides during the cyanide leaching process. The suitable temperature for oxidizing roasting of carbonaceous gold ores ranges from 650 to 700 °C, and a roasting duration of 1.5 to 2 h is necessary to achieve a high gold leaching rate during subsequent cyanidation [15]. Chemical oxidation is a pretreatment method that involves adding oxidizing agents to oxidize carbonaceous gold ores. Commonly used oxidants include chlorine gas, sodium chlorate, sodium hypochlorite, ozone, hydrogen peroxide, potassium permanganate, and nitric acid [16]. For instance, introducing chlorine gas into the pulp generates hypochlorous acid through reaction with water, which passivates the surface of the carbonaceous material, thereby reducing its preg-robbing capacity. Simultaneously, sulfur in sulfides is oxidized, exposing the encapsulated gold. This method demonstrates promising potential for industrial application, though it also presents challenges such as high economic costs and environmental concerns [17]. Furthermore, due to the excellent floatability of both carbonaceous matter and sulfide minerals, when gold is primarily encapsulated within them, a preferential flotation method can be employed to float these components. After discarding a large amount of tailings, the gold concentrate undergoes oxidizing roasting to remove carbon and sulfur, followed by cyanide leaching for gold extraction [18]. When the gold-bearing minerals are not carbonaceous matter or sulfides, the carbonaceous matter and sulfides are pre-removed by flotation, and the flotation tailings are then subjected to oxidizing roasting before leaching. The flotation method effectively enriches gold, significantly reduces the processing load for subsequent roasting and leaching operations, and lowers production costs. In recent years, many researchers have explored pretreatment and leaching processes for carbonaceous gold ores. Jin [15] applied oxidative roasting (650 °C, 1.5 h) to fine-grained carbonaceous gold ore and achieved a gold leaching rate of 89.6% during subsequent cyanidation. Guo [19] used chemical oxidation pretreatment with a chlorine-hypochlorite solution, obtaining a gold leaching rate of approximately 85%. Xu [20] developed a nickel-catalyzed ammoniacal thiosulfate leaching-resin adsorption process, achieving a gold leaching rate of 92%. In comparison, the present study adopts a combined flotation pre-concentration, oxidative roasting, and Mill–Leach Coordination (MCL) process. Under the conditions of roasting at 650 °C for 1 h, followed by MCL leaching with a “Jincan” dosage of 3 kg/t for 16 h, a gold leaching rate of 97.12% is achieved. This process offers advantages in reagent consumption and leaching time. Moreover, the kinetic analysis of the MCL process reveals an internal diffusion control mechanism, providing a theoretical basis for process optimization.

Although the flotation–roasting–cyanidation process for carbonaceous gold ores is well established, this study employs the Mill–Leach Coordination (MCL) process (originally developed for zinc oxide ore, see Ref. [21]) to enhance the leaching of roasted concentrate

from high-carbon gold ore and improve Au recovery. On this basis, an empirical kinetic equation under simultaneous grinding and leaching conditions is derived. Unlike conventional leaching, the MCL process continuously renews the particle surface and eliminates product-layer diffusion resistance, thereby achieving a significantly higher gold leaching rate in a shorter time. The kinetic model and parameters obtained in this work are original and can provide quantitative guidance for industrial application.

2. Experiment and Methods

2.1. Sample Source and Preparation

Test Sample: The test sample was obtained from a gold ore deposit in Laos. Representative ore samples totaling 1 ton were collected through multi-point field sampling. These samples underwent processing steps including crushing (using a jaw crusher and sieve shaker, both from Retsch GmbH, Haan, Germany), screening, mixing, and division to obtain a sample with a particle size of -2 mm, which was used for subsequent experimental research.

Process Mineralogy Samples: Based on analytical requirements, lump ore samples were manually selected and polished into thin sections and polished blocks (using an automatic grinding/polishing machine from Struers ApS, Ballerup, Denmark) for microscopic examination. A bulk composite sample was mixed, homogenized, and divided; 500 g of this sample was ground to a particle size of -74 μm (using a planetary ball mill from Fritsch GmbH, Idar-Oberstein, Germany) for chemical multi-element analysis and X-ray diffraction (XRD) analysis. XRD analysis was performed on a D8 Advance diffractometer (Bruker AXS GmbH, Karlsruhe, Germany) with the Diffrac.Suite software (version v5.0) for phase identification. Another 100 g were ground to -300 mesh (approximately 48 μm), then mixed, divided, and resin-mounted for Mineral Liberation Analysis (MLA) and electron probe microanalysis (EPMA). MLA was carried out using an MLA system attached to a Quanta 650 scanning electron microscope (Thermo Fisher Scientific, Hillsboro, OR, USA), with data acquisition and processing using MLA Suite software (version v3.1, JKTech Pty Ltd., Brisbane, QLD, Australia). EPMA was conducted on a JXA-8230 electron probe microanalyzer (JEOL Ltd., Akishima, Tokyo, Japan), with its accompanying data processing software (version v1.2) for quantitative correction. An additional 2 kg of the sample were ground to -150 μm for separate single-mineral analysis.

2.2. Experimental Methods

Flotation Test: 500 g of the ore sample was weighed and ground to a specific particle size. It was then placed in the flotation cell of a mechanical flotation machine. Reagents including an activator, collector, dispersant, and frother were added sequentially. After aeration, the flotation scraper was activated to produce a concentrate.

Leaching Test: This study employed a Mill–Leach Coordination (MCL) process experiment; a schematic diagram of the equipment is shown in Figure 1 [21]. 300 g of the raw leaching sample was weighed and added to the reaction vessel. A stirrer was used to agitate and disperse the solution and mineral powder within the container, ensuring effective mixing. Upon completion of the leaching process, the mixture was filtered, and the Au content in both the leachate and the leach residue was analyzed. All leaching experiments were conducted in triplicate, and all results are presented as averages.

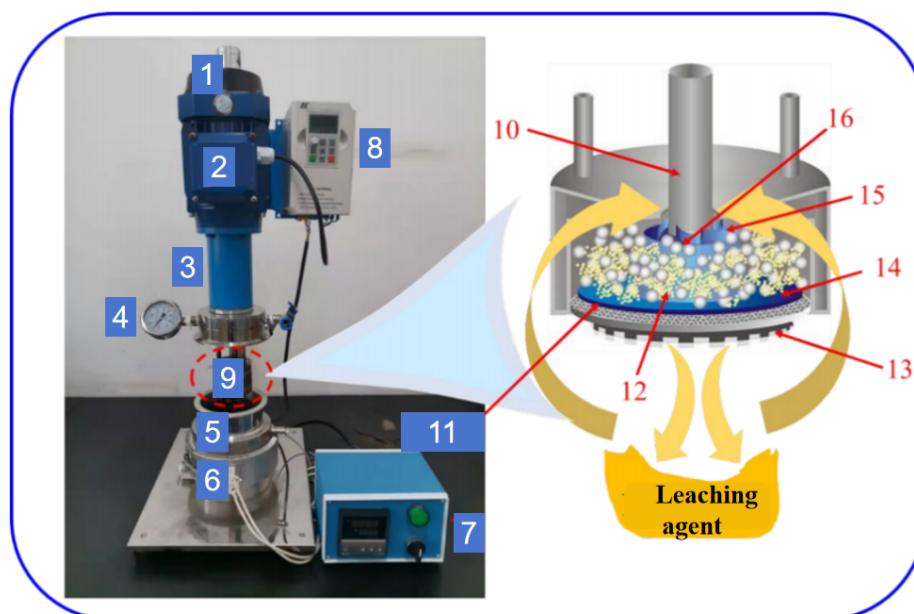


Figure 1. Schematic diagram of the reaction apparatus (1—Heat dissipation fan; 2—Motor; 3—Rotating shaft; 4—Vacuum pressure gauge; 5—Reaction cylinder; 6—Electric heating jacket; 7—Temperature control device; 8—Grinding control system; 9—Grinding basket; 10—Drive shaft; 11—Separator; 12—Raw ore powder; 13—Agitator; 14—Grinding basket chamber; 15—Slurry pump wheel; 16—Grinding media).

2.3. Experimental Flowsheet

The industrial production flowsheet used on-site is shown in Figure 2. It involves gravity separation using a Knelson concentrator, followed by leaching of the gravity separation tailings. The traditional process achieved a gold leaching efficiency exceeding 90%. However, due to changes in the raw ore properties, notably a significant increase in carbon content, the original process became inadequate. The results of a systematic comparison of different processes are shown in Table 1. The leaching agent referred to as “Jincan” is a commercially available, environmentally friendly gold leaching reagent. Its core active component is sodium tricyanomelaminates ($C_6H_3N_6Na_3O_3$), which contains cyano groups (-CN) capable of complexing and solubilizing gold from the ore. The reagent is formulated with auxiliary ingredients including alkaline polymeric iron, alkalis, carbonates, and small amounts of complexing agents and protective agents. The design principle is to provide leaching efficiency comparable to that of sodium cyanide while fundamentally reducing toxicity through covalent bonding that locks the free cyanide ions.

As shown in the table, the direct leaching process resulted in a low gold leaching rate with significant losses. Furthermore, the leaching agent “Jincan” proved more effective than NaCN. Although Jincan achieved a slightly higher leaching rate than NaCN (ca. 2.5 percentage points), given its environmental advantages, Jincan can be considered a feasible alternative to NaCN. After the raw ore was roasted, the gold leaching rate under identical conditions increased significantly, exceeding 90%, indicating high leaching efficiency. However, the low gold grade of the raw ore means direct roasting involves processing large volumes of material, leading to high energy consumption and costs. Based on preliminary exploratory tests, a combined Flotation–Roasting–Grinding–Leaching Synergy (MCL) process is proposed. The flowsheet for this process is shown in Figure 3.

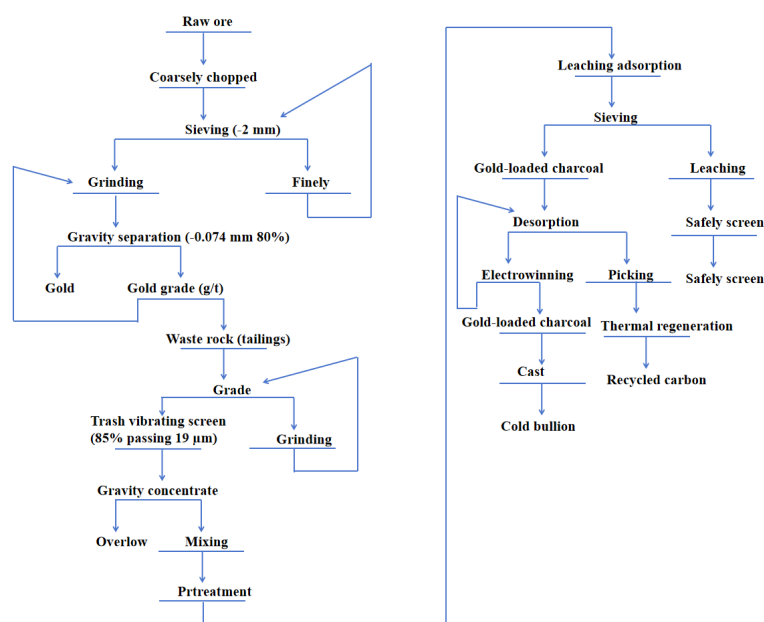


Figure 2. Flowsheet of on-site gold extraction.

Table 1. Comparison of leaching rates under different processes.

Process 50.	Reaction Conditions	Au Leaching Rate (%)
Direct Leaching	Room temperature, Leaching agent: NaCN 50 kg/t, Leaching time: 48 h, Particle size: 85% passing 19 μm, L/S ratio: 6 mL/g	52.31%
Direct Leaching	Room temperature, Leaching agent: Jincan 50 kg/t, Leaching time: 48 h, Particle size: 85% passing 19 μm, L/S ratio: 6 mL/g	60.35%
Roasting–Leaching	Raw ore roasted at 600 °C for 1.5 h, Leaching agent: NaCN 50 kg/t, Leaching time: 48 h, Particle size: 85% passing 19 μm, L/S ratio: 6 mL/g	90.43%
Roasting–Leaching	Raw ore roasted at 600 °C for 1.5 h, Leaching agent: Jincan 50 kg/t, Leaching time: 48 h, Particle size: 85% passing 19 μm, L/S ratio: 6 mL/g	92.86%

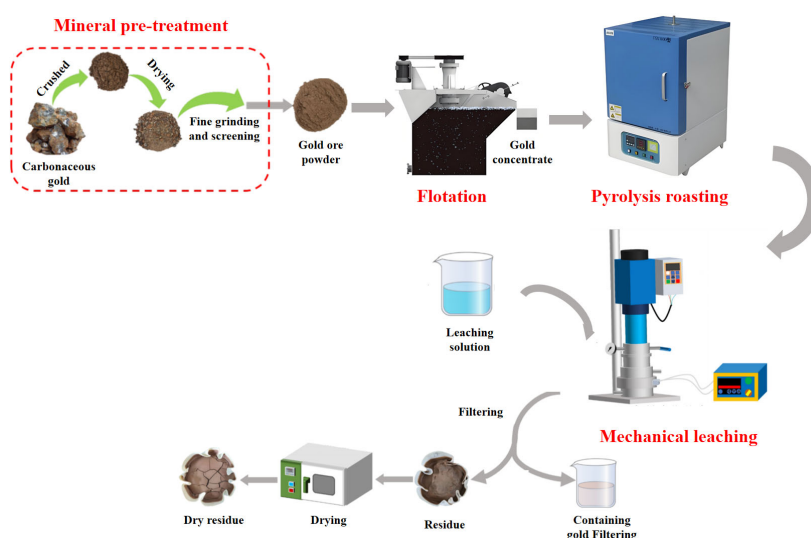


Figure 3. Test Process Flow.

3. Results and Discussion

3.1. Process Mineralogy Study of the Ore

3.1.1. Chemical Element Analysis of the Ore Sample

The chemical composition analysis of the carbonaceous gold ore is presented in Table 2. As shown, the contents of Au and Ag are 6.31 g/t and 15.68 g/t, respectively, with Au possessing a significant recovery value. The harmful elements affecting gold recovery, namely carbon and sulfur, are present in high concentrations of 5.27% and 1.43%, respectively, classifying it as a refractory high-carbon gold ore. Additionally, the content of gangue minerals such as Si, Fe, Al, and Mg is high.

Table 2. Chemical multi-element analysis of the gold ore.

Element	Au *	Ag *	S	CaO	MgO	Al ₂ O ₃
Content (%)	6.31	15.68	1.43	0.14	0.22	9.21
Element	Na ₂ O	Fe	SiO ₂	As	Cu	C _{Total}
Content (%)	0.061	7.74	70.60	0.51	0.029	5.27

* Units for Au and Ag are g/t.

The phase analysis of Au and C in the raw ore was conducted, and the results are shown in Tables 3 and 4. Using chemical phase analysis through sequential selective dissolution (first cyanide leaching for liberated gold, then hydrofluoric acid for silicates, hydrochloric acid for carbonates, and nitric acid for sulfides) to determine the gold content in each phase, thereby distinguishing liberated gold from gold encapsulated in different minerals. The results indicate that the encapsulation rate of Au in the ore is as high as 83.68%, with only 16.32% of the Au occurring as liberated particles. The contents of organic carbon and graphitic carbon in the raw ore are 1.76% and 2.87%, respectively. Direct leaching leads to a severe “preg-robbing” effect, resulting in a low leaching rate.

Table 3. Chemical phase analysis of Au in the raw ore.

Phase	Liberated Au	Au Encapsulated in Silicates	Au Encapsulated in Carbonates	Au Encapsulated in Sulfides	Total
Content (g/t)	1.03	3.42	1.07	0.79	6.31
Distribution (%)	16.32	54.20	16.95	12.52	100

Table 4. Chemical phase analysis of carbon in the raw ore.

Phase	Organic Carbon	Graphitic Carbon	Carbonates	Total
Content (g/t)	1.76	2.87	0.64	5.27
Distribution (%)	33.40	54.46	12.14	100

3.1.2. Mineral Composition Analysis of the Ore

Mineral liberation analysis (MLA) was conducted to analyze and quantify the mineral particles in the ore to determine its mineral composition. The MLA pattern is shown in Figure 4, and the results for part of the mineral composition and content are presented in Table 5. As shown in the table, the ore comprises 18 different mineral species. The only independent gold mineral is native gold. Metallic minerals include pyrite and minor amounts of arsenopyrite, chalcopyrite, and sphalerite. The gangue minerals are predominantly quartz, plagioclase, sericite (muscovite), and ankerite, accounting for approximately 93% of the total.

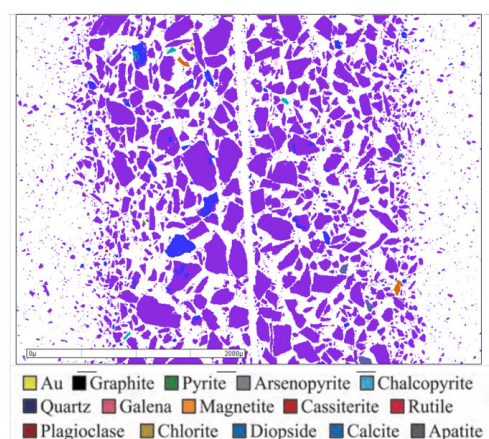


Figure 4. General MLA Diagram of Ore.

Table 5. Mineral Composition and Content.

Minerals	Molecular Formula	Content	Minerals	Molecular Formula	Content
Natural gold	Au	0.0006325	Potassium feldspar	$\text{KAl}_2[\text{AlSi}_3\text{O}_{10}][\text{OH}]_2$	17.8
Sphalerite	ZnS	0.05	Kaolinite	$\text{Al}_4[\text{Si}_4\text{O}_{10}](\text{OH})_8$	0.3
Pyrite	FeS_2	2.6	Zircon	ZrSiO_4	0.01
Orpiment	As_2S_3	1.11	Calcite	$\text{Ca}[\text{CO}_3]$	0.2
Realgar	As_4S_4	0.02	Dolomite	$\text{CaMg}[\text{CO}_3]_2$	0.5
Quartz	SiO_2	56.42	Fluorite	CaF_2	1.23
Goethite	FeOOH	12.90	Scorodite	$\text{Fe}[\text{AsO}_4] \cdot 2\text{H}_2\text{O}$	0.03
Talc	$\text{Mg}_3[\text{Si}_4\text{O}_{10}](\text{OH})_2$	2.45	Scheelite	CaWO_4	0.02
Hornblende	$(\text{Mg,Fe,Al})_3(\text{OH})_6\{(\text{Mg,Fe,Al})_3[(\text{Si,Al})_6\text{O}_{10}(\text{OH})_2]\}$	1.3	C	C	5.21

3.1.3. Occurrence and Size Distribution of Au

X-ray energy dispersive spectroscopy (EDS) analysis was randomly performed on 11 particles of native gold; the results are shown in Figure 5. The analysis indicated that the native gold contained 99.28% Au, 0.30% Ca, and 0.42% Fe. Through ore microscopy and MLA, the particle size of 130 native gold particles was measured. The measurement results revealed that the native gold particles were primarily distributed within the size ranges of $-19 + 9.5 \mu\text{m}$ and $-9.5 + 4.75 \mu\text{m}$, with distribution rates of 65.42% and 28.93%, respectively. The gold particles in the ore are relatively fine, with the vast majority classified as micro-sized gold. Consequently, achieving effective enrichment using Knelson gravity separation is challenging, necessitating an improvement in the original beneficiation process.

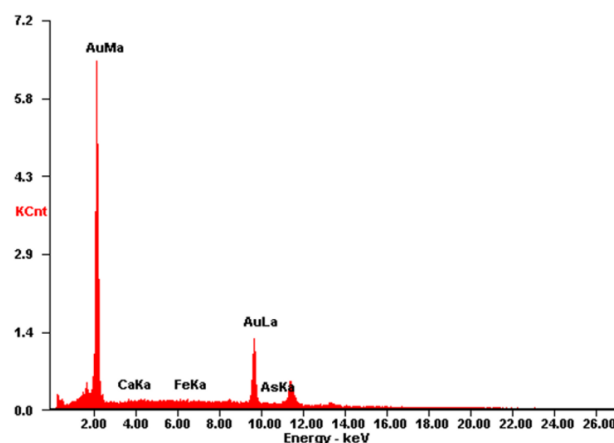


Figure 5. Natural gold X-ray energy spectrum.

Observation under ore microscopy revealed that the native gold particles exhibit irregular, rounded, scaly, dendritic, and other anhedral forms. The encapsulated gold primarily occurs as irregular or rounded particles enclosed within calcite and quartz, with a very small fraction encapsulated within pyrite. Occasional instances were observed where gold is distributed within fractures of quartz or intergrown with orpiment and fluorite, as specifically illustrated in Figure 6.

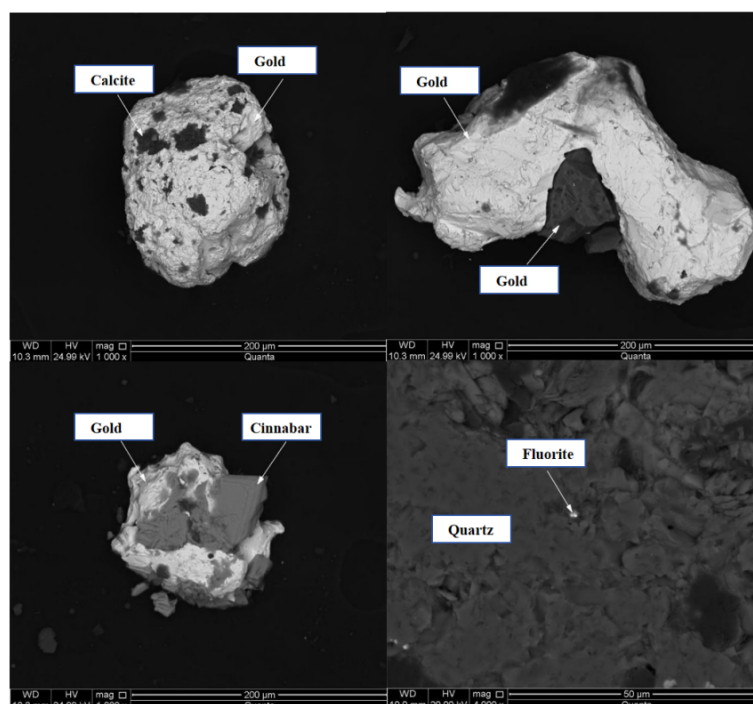


Figure 6. Inlaid characteristics of natural gold.

3.2. Pre-Concentration of Gold by Flotation

Based on the mineralogical analysis and preliminary exploratory tests, a combined process of flotation pre-concentration, oxidative roasting, and leaching was proposed to treat this carbonaceous gold ore. To investigate the effects of various flotation parameters, including grinding fineness, type of activator, type of collector, and type of depressant, tests were conducted according to the flowsheet shown in Figure 7.

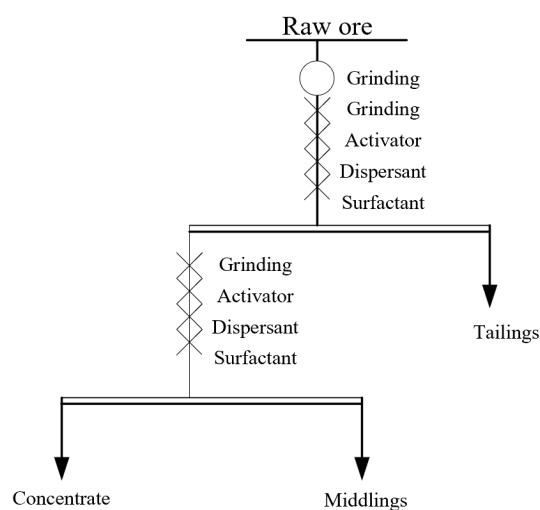


Figure 7. Flotation test process flow.

3.2.1. Influence of Different Flotation Parameters

The effect of grinding fineness on gold flotation performance is shown in Figure 8a. As the proportion of $-74\ \mu\text{m}$ particles increased, both the grade and recovery of Au in the concentrate initially increased and then decreased. When the percentage of $-74\ \mu\text{m}$ particles in the ground feed was between 75% and 85%, both grade and recovery remained relatively high. At 80% passing $74\ \mu\text{m}$, the gold grade and recovery reached 19.32 g/t and 93.26%, respectively.

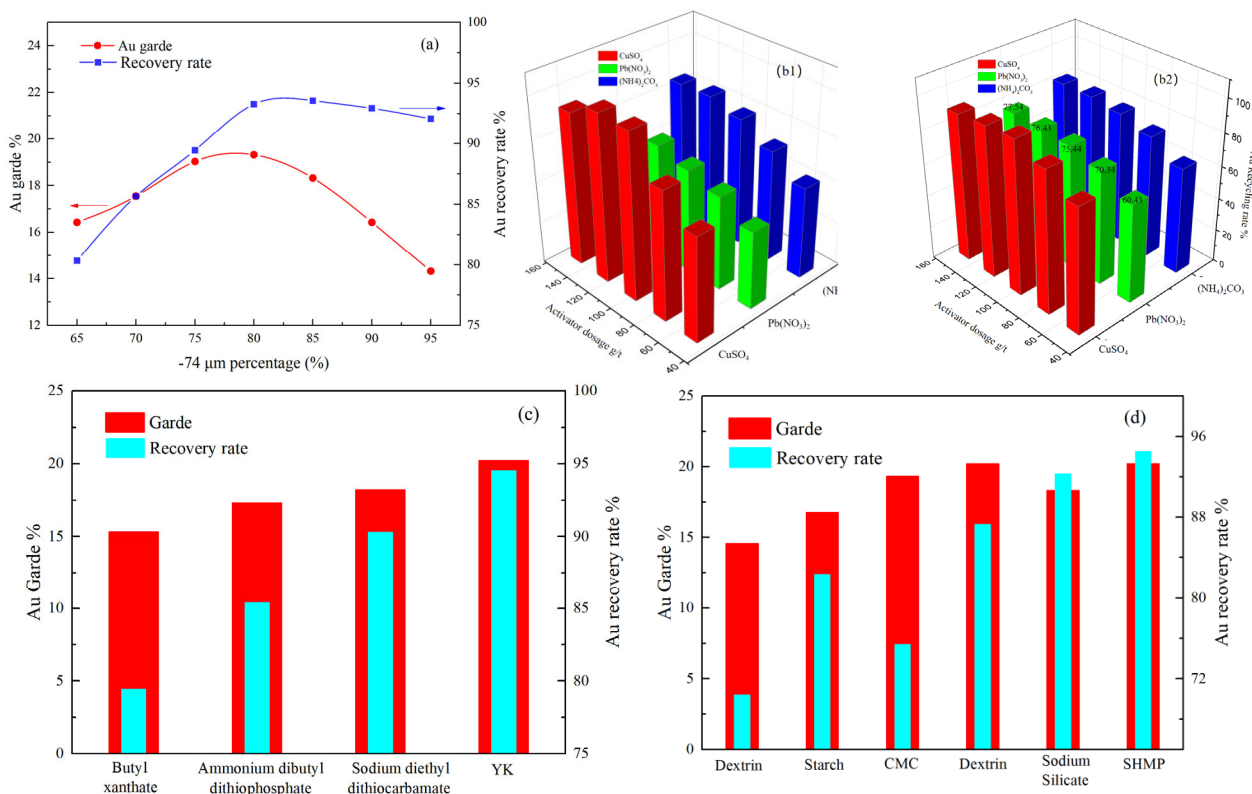


Figure 8. Grade and recovery rate of gold concentrate under different test conditions ((a) Influence of grinding fineness; (b1) Influence of activator on grade; (b2) Influence of activator on recovery rate; (c) Influence of collector; (d) Influence of dispersant).

Activators in gold flotation can promote the formation of a hydrophobic film on the mineral surface, enhancing the adsorption of collectors. Tests compared the effects of different activators, including CuSO₄, Pb(NO₃)₂, and (NH₄)₂CO₃, on gold flotation; the results are shown in Figure 8(b1,b2). The effectiveness of the activators followed the order: CuSO₄ > (NH₄)₂CO₃ > Pb(NO₃)₂. The recovery initially increased and then decreased with increasing dosage of CuSO₄. At a CuSO₄ dosage of 100 g/t, the gold grade and recovery were 19.87 g/t and 94.04%, respectively. Copper sulfate acts as an activator to activate the surface of gold minerals and pyrite, promoting collector adsorption.

Tests compared the effects of different collectors, including sodium butyl xanthate, ammonium dibutyl dithiophosphate, diethyldithiocarbamate, and a novel collector YK, which are used to enhance the hydrophobicity of gold and sulfide minerals. The results are shown in Figure 8c. The novel collector YK demonstrated the best gold collection performance, making it more suitable for the flotation recovery of this type of gold ore.

Tests compared the effects of different depressants, including dextrin, starch, carboxymethyl cellulose (CMC), sodium silicate, and sodium hexametaphosphate (SHMP), which are depressants used to inhibit the flotation of gangue minerals (e.g., quartz, calcite, sericite). The results are shown in Figure 8d. Sodium hexametaphosphate provided the

best depression effect on gangue minerals. At a dosage of 200 g/t, the grade and recovery of the gold concentrate were 20.22 g/t and 94.54%, respectively.

3.2.2. Closed-Circuit Tests

The flotation flowsheet structure (one roughing, two cleaning, and two scavenging stages) was optimized based on the results of preliminary conditional tests, by comparing the closed-circuit test results of different flowsheet configurations. The flowsheet and reagent dosages are shown in Figure 9. The results of the closed-circuit tests are presented in Table 6. A final gold concentrate with a grade of 21.03 g/t and a recovery of 96.57% was achieved. The carbon content in the concentrate reached 4.51%.

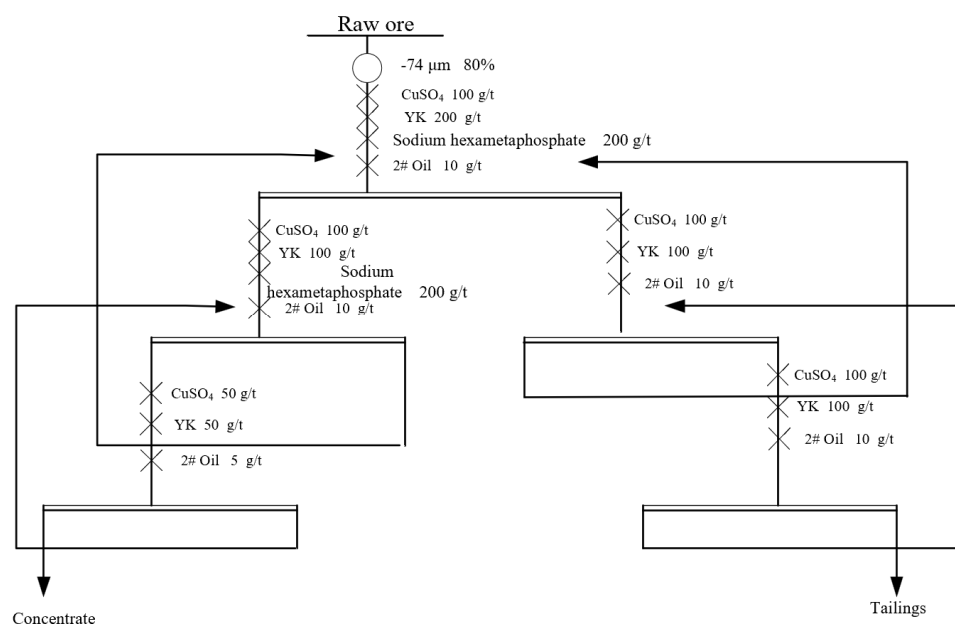


Figure 9. Closed-loop Process Flow.

Table 6. Results of the Flotation Closed-Circuit Test.

Product	Yield/%	Gold Grade (g/t)	Gold Recovery Rate (%)
Gold concentrate	28.97	21.03	96.57
Tailings	71.03	0.305	3.43
Raw ore	100.00	6.31	100.00

3.3. Oxidative Roasting

The concentrate produced from the closed-circuit flotation process still contained a high carbon content. Therefore, it was subjected to high-temperature roasting in a muffle furnace to remove organic carbon and mitigate its adverse impact on the subsequent leaching process. To address the environmental risk posed by volatile arsenic compounds from arsenic-bearing minerals in the concentrate, a small amount of lime (CaO) was added to the concentrate prior to roasting to immobilize arsenic as non-volatile calcium arsenate. Furthermore, the off-gas generated during muffle furnace roasting was passed through a series of alkaline absorption bottles (NaOH solution) and an activated carbon trap before being released into the fume hood. The roasted product was leached under the following conditions: pH = 11–12, leaching agent “Jincan” dosage of 2 kg/t, liquid-to-solid ratio of 6:1 mL/g, stirring speed of 350 rpm, at room temperature for 24 h. The effects of different roasting temperatures and durations on the gold leaching rate were compared; the results are shown in Figure 10. As illustrated in the figure, the gold leaching rate increased initially

and then plateaued with increasing roasting temperature. The flotation concentrate roasted at 650 °C exhibited the highest gold leaching rate (87.31%) during subsequent cyanidation. Similarly, the leaching rate increased initially and then decreased with extended roasting time. The optimal leaching performance was obtained after a roasting duration of 1 h.

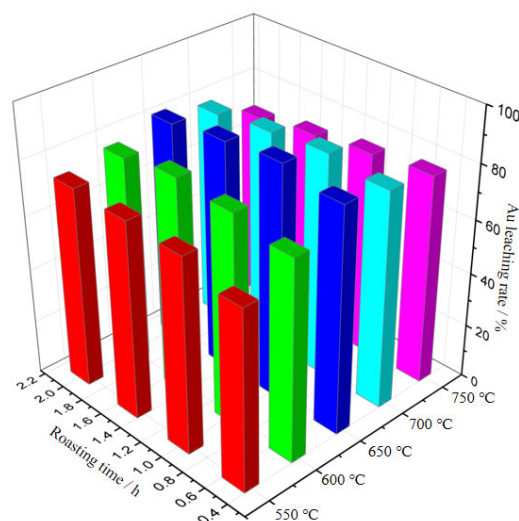


Figure 10. Influence of different calcination temperatures and times on the gold leaching rate.

3.4. Grinding-Leaching Synergistic (MCL) Process

The direct leaching process resulted in a gold leaching rate of less than 90%. A basket mill was employed as mechanical activation equipment. Zirconia cylinders ($\Phi 2$ mm) were added to the reactor. The leaching efficiency of the Grinding-Leaching Synergistic (MCL) process was compared with that of the Conventional Stirred Leaching (CSL) process. The effects of leaching time, leaching agent concentration, reaction pH, and stirring speed were investigated.

Effect of Leaching Time: Under the conditions of a leaching agent (“Jincan”) dosage of 2 kg/t, pH = 11, and a stirring speed of 350 rpm, the influence of leaching time on the Au leaching rate was studied. The results are shown in Figure 11a. The Au leaching rate for the MCL process essentially reached equilibrium at 16 h, achieving a rate of 95.78%. In contrast, the CSL process required approximately 24 h to achieve near-complete Au dissolution, with a final leaching rate of 90.32%.

Effect of Leaching Agent Concentration: Under the conditions of pH = 11, a stirring speed of 350 rpm, and a leaching time of 24 h, the effect of “Jincan” concentration on the Au leaching rate was studied. The results are shown in Figure 11b. The Au leaching rate increased rapidly with increasing leaching agent dosage, rising from 73.45% to 96.79%. The leaching rate stabilized when the leaching agent dosage reached 3 kg/t.

Effect of pH: Under the conditions of a leaching agent dosage of 3 kg/t, a stirring speed of 350 rpm, and a leaching time of 24 h, the influence of slurry pH on the Au leaching rate was studied. The results are shown in Figure 11c. The Au leaching rate initially increased and then decreased with increasing pH. The maximum leaching rate of 96.82% was achieved at pH = 11.

Effect of Stirring Speed: Under the conditions of a leaching agent dosage of 3 kg/t, pH = 11, and a leaching time of 24 h, the effect of stirring speed on the Au leaching rate was studied. The results are shown in Figure 11d. Increasing the stirring speed helped eliminate external diffusion resistance, promoting the complete diffusion of gold into the leaching solution and thereby enhancing leaching efficiency. However, further increases in stirring speed beyond 400 rpm did not significantly improve leaching performance but increased

energy consumption. Therefore, a stirring speed of 400 rpm was selected, achieving a Au leaching rate of 97.12%.

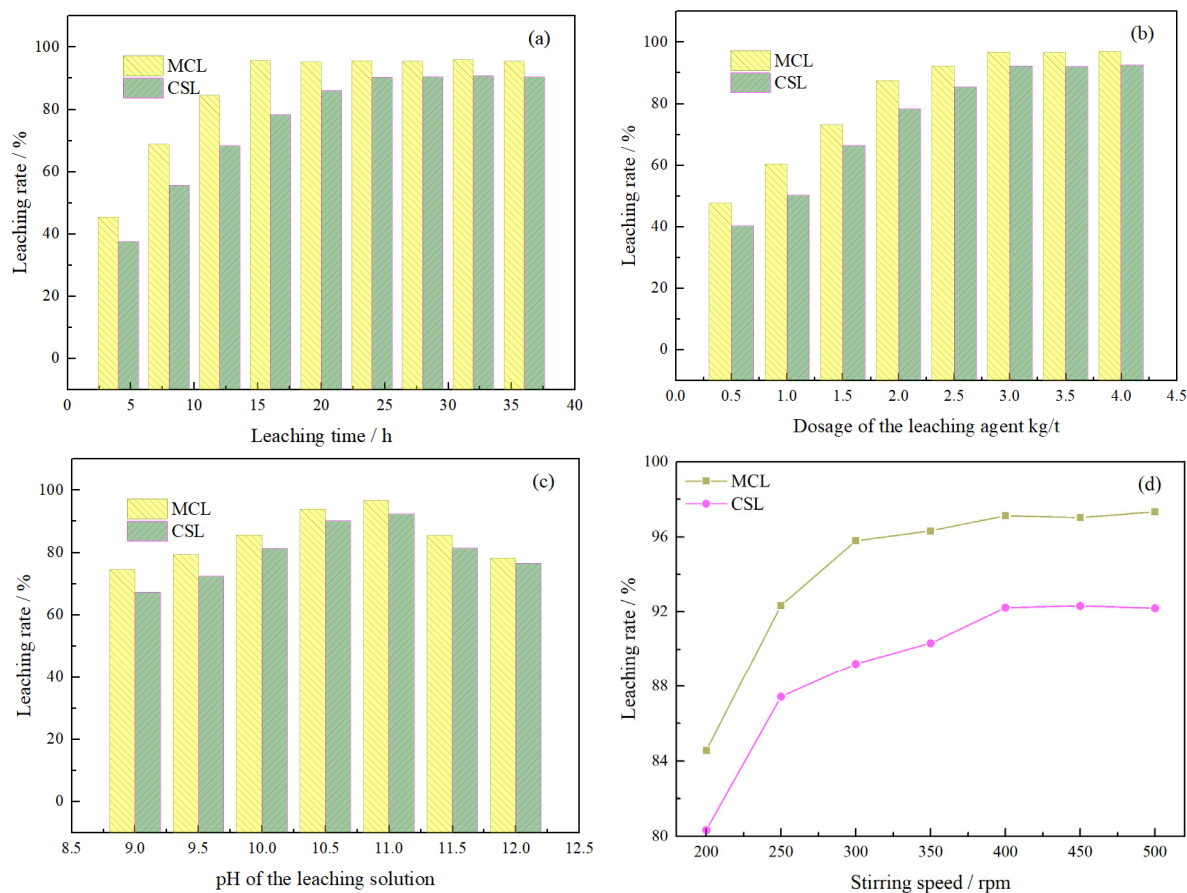


Figure 11. Influence of MCL and CSL processes on the gold leaching rate under different leaching conditions ((a) Leaching time; (b) Leaching agent dosage; (c) pH of leaching solution; (d) Stirring rate).

3.5. Leaching Kinetics

Based on the Au leaching results from the MCL process, the leaching kinetics within this system were investigated to identify the most appropriate kinetic model and equations, thereby providing a theoretical foundation for the resource utilization of this gold ore.

3.5.1. Leaching Model

From the perspective of conventional leaching processes, the rate-limiting step is generally attributed to the external/internal diffusion of the leaching agent, the chemical reaction at the surface, or the internal/external diffusion of soluble products. The leaching process often follows the unreacted shrinking core model. However, in a simultaneous grinding-leaching process, the particle size continuously decreases due to the grinding action of the media, exhibiting a shrinking trend that does not strictly conform to the unreacted shrinking core model but rather aligns better with a shrinking core-shrinking particle model. A significant challenge in applying this model is the difficulty in measuring the thickness of the product layer. Furthermore, since particle size changes continuously during the grinding-leaching process, identifying a more suitable kinetic model is necessary.

Based on previous research, the Avrami equation, a semi-empirical model, is often used to describe the reaction kinetics of leaching processes for various metals and metal oxides [22–25]. The equation is expressed as follows:

$$-\ln(1 - \alpha) = kt^n \quad (1)$$

It can also be transformed logarithmically into:

$$\ln[-\ln(1 - \alpha)] = \ln k + n \ln t \quad (2)$$

where α is the gold leaching rate, k is the apparent rate constant of the leaching reaction, and n is a constant determined by the type of leaching reaction. The value of n indicates the rate-controlling mechanism: values close to or equal to 1 suggest chemical reaction control; values within the range (0, 0.5) indicate diffusion control; and values between (0.5, 1) represent mixed control by both mechanisms.

The data of leaching rate versus reaction time were fitted to establish the relationship between $\ln[-\ln(1 - \alpha)]$ and $\ln t$, as shown in Figure 12. A strong linear correlation between $\ln[-\ln(1 - \alpha)]$ and $\ln t$ was observed, indicating that the gold leaching kinetics under the MCL process conform to the Avrami model. Furthermore, the calculated n values of the gold leaching equations at different temperatures were relatively close to each other and all less than 0.5. This suggests that the gold leaching process via the MCL method is consistent with a diffusion-controlled mechanism. Additionally, the rate of gold leaching exhibited a decelerating trend over time.

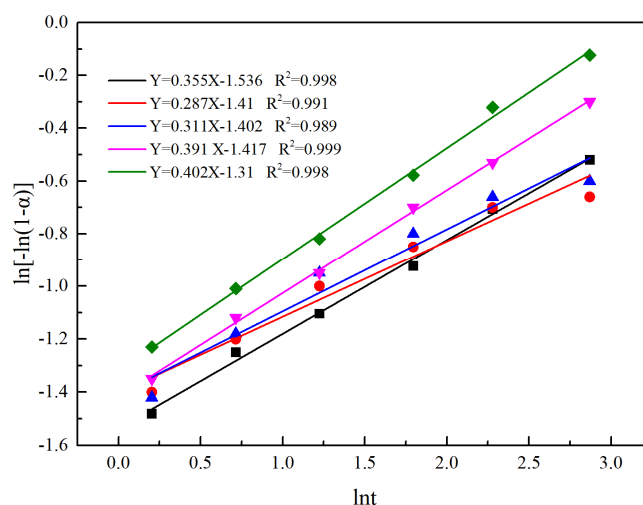


Figure 12. Linear fitting of Au leaching rate at different temperatures.

3.5.2. Effect of Temperature on Gold Leaching

The gold leaching rate data at different temperatures and the fitted “ n ” values were substituted into Equation (1), yielding the fitting relationship between $-\ln(1 - \alpha)$ and $t^{0.349}$, as shown in Figure 13a. The slopes of the different fitting curves in Figure 13a represent the apparent rate constants (k) at their respective temperatures. The apparent activation energy for the gold leaching process can be calculated using the Arrhenius equation:

$$k = A \times \exp(-E_a/RT) \quad (3)$$

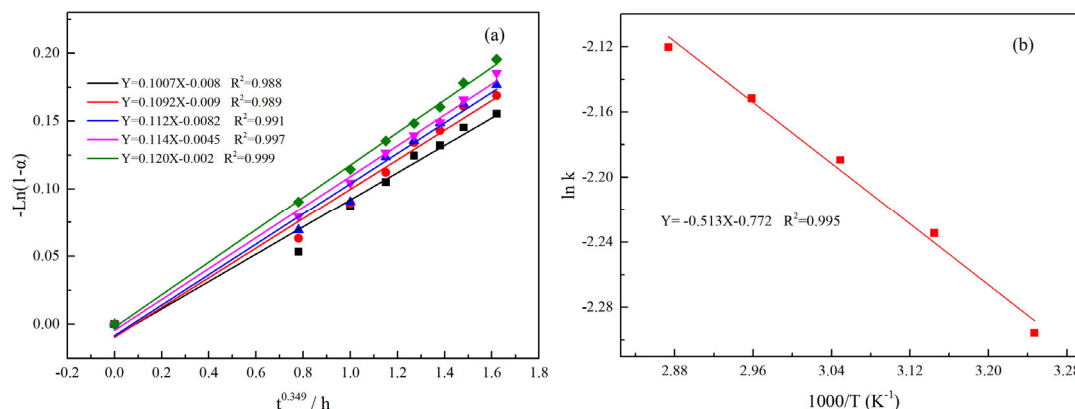


Figure 13. (a) Linear fitting of Au leaching rate; (b) Relationship graph between $\ln k$ and $1000/T$ for Au leaching.

Taking the natural logarithm of both sides gives:

$$\ln k = \ln A - E_a/RT \quad (4)$$

where E_a is the apparent activation energy (kJ/mol), A is the pre-exponential factor, k is the apparent rate constant, R is the gas constant ($8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$), and T is the absolute leaching temperature (K).

The plot of $\ln k$ versus $1000/T$ is shown in Figure 13b. The slope of the resulting linear fit is equal to $-E_a/(1000R)$, from which the apparent activation energy for the gold leaching process via the MCL method was calculated to be 4.27 kJ/mol. The magnitude of the apparent activation energy can be used to further identify the rate-limiting step of the leaching reaction. Since the activation energy falls within the range of 4.2–12 kJ/mol, it can be concluded that the reaction is controlled by internal diffusion [26].

3.5.3. Influence of Various Factors on the Leaching Rate

In summary, the Avrami equation can accurately describe the gold leaching behavior under the MCL process, with the rate-controlling step being internal diffusion control. Similarly, to further investigate the relationship between the gold leaching rate and factors such as leaching agent concentration, liquid-to-solid ratio, and stirring speed, the following semi-empirical model was established, as shown in Equation (5):

$$-\ln(1-\alpha) = k_0 C_A^a (L/S)^b (\text{rpm}) \exp(-E_a/RT) t^n \quad (5)$$

where α is the gold leaching rate, k_0 is the pre-exponential factor, C_A is the leaching agent concentration (kg/t), L/S is the liquid-to-solid ratio (mL/g), rpm is the stirring speed (r/min), E_a is the apparent activation energy (4.27 kJ/mol), R is the gas constant ($8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$), T is the absolute temperature (K), t is the leaching time (h), and n is the Avrami exponent (0.394).

Based on the controlled variable method, the reaction orders for different factors were derived, as detailed in the corresponding figures. Figure 14a–c show the relationship between $-\ln(1-\alpha)$ and $t^{0.349}$ under different conditions of leaching agent concentration, liquid-to-solid ratio, and stirring speed. The slopes of the fitted lines represent the reaction rate constant k . Linear relationships between $\ln k$ and $\ln C_A$, $\ln(L/S)$, and $\ln(\text{rpm})$ were then established. The fitted reaction orders were 0.712, 1.78, and 2.62, respectively. The magnitude of these reaction orders reflects the degree of influence each factor has on the gold leaching rate.

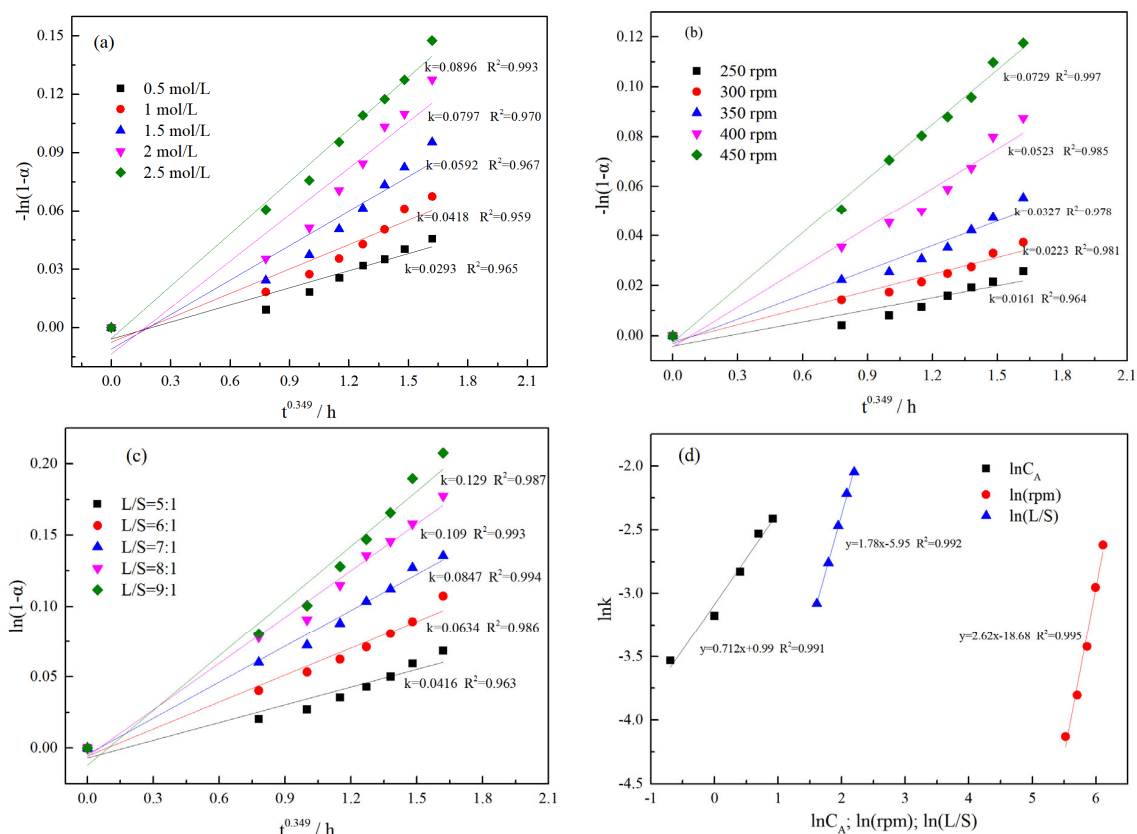


Figure 14. (a) The influence of leaching agent concentration on leaching rate; (b) The influence of stirring rate on leaching rate; (c) The effect of L/S on leaching rate; (d) Linear fitting of $\ln k$ with $\ln C_A$, $\ln(L/S)$, and $\ln(\text{rpm})$.

The derived reaction orders were substituted into the leaching kinetic Equation (5), and the relationship between $-\ln(1 - \alpha)$ and $k_0 C_A^a (L/S)^b (\text{rpm})^c \exp(-E_a/RT) t^n$ was plotted, as shown in Figure 15. The graph shows that the gold leaching results under different conditions are evenly distributed on both sides of the straight line, the slope of which is 0.0146. Therefore, the kinetic equation for gold leaching via the MCL process can be expressed as Equation (6):

$$-\ln(1 - \alpha) = 1.46 \times 10^{-2} C_A^{0.712} (L/S)^{1.78} (\text{rpm})^{2.62} \exp(-4270/RT) t^{0.394} \quad (6)$$

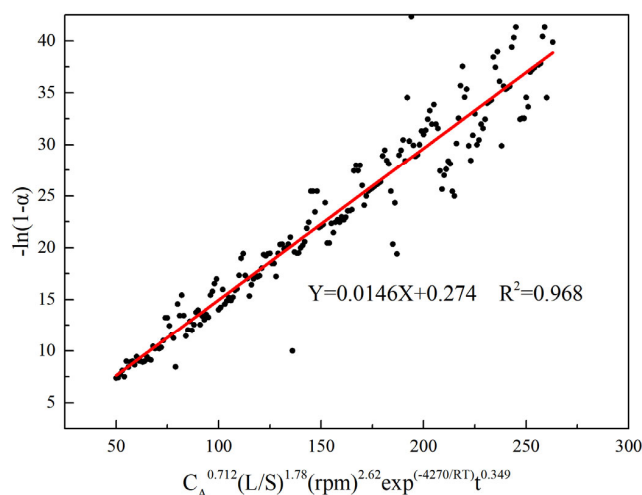


Figure 15. Graph of $-\ln(1 - \alpha)$ of Au leaching rate versus $k_0 C_A^a (L/S)^b (\text{rpm})^c \exp(-E_a/RT) t^n$.

4. Conclusions

- (1) The high-carbon gold ore from Laos is characterized by fine-grained, encapsulated gold and high contents of organic carbon and graphitic carbon, which are the primary causes of its poor direct leaching performance.
- (2) A flotation flowsheet consisting of one roughing, two cleaning, and two scavenging stages effectively pre-concentrates gold and removes a large amount of gangue minerals, significantly reducing the feed volume for subsequent roasting and leaching operations.
- (3) Oxidative roasting effectively removes organic carbon from the flotation concentrate. The Mill–Leach Coordination (MCL) process, which integrates simultaneous grinding and leaching, achieves substantially higher gold extraction efficiency than conventional stirred leaching, owing to continuous surface renewal and elimination of product-layer diffusion resistance.
- (4) The leaching kinetics under the MCL process conform to the Avrami model and are controlled by internal diffusion. The derived semi-empirical kinetic equation provides a theoretical basis for process optimization and industrial scale-up. The proposed combined flotation–roasting–MCL process offers a promising and referenceable technical route for the efficient utilization of similar refractory high-carbon gold ores.

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