



# Article Strengthening Gold Extraction from Carbonaceous Gold Ore Based on Decarburization by Two-Stage Fluidized Oxidation Roasting

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**Abstract:** Oxidation roasting is one of the most effective methods to remove carbonaceous matter from carbonaceous gold ore. However, the consolidation of calcine is a common problem caused by a large amount of heat release during the oxidation roasting of gold ore with high carbon and sulfur content. In this study, a two-stage fluidized oxidation roasting method was developed to strengthen gold extraction. Pre-roasting was performed in a low oxygen environment for the mild combustion of carbonaceous matter, and then the pre-roasted product was subjected to secondary roasting followed by leaching. The results show that reducing the carbon content in the ore by pre-roasting can effectively lower the peak temperature inside the material. After the second-stage roasting, many cracks and pores were formed in silicates and carbonates, thus increasing the distribution of exposed gold from 28.95% to 88.01%. Finally, the extraction of gold increased from 4.55% to 86.10%. Overall, this study provides references for an efficient strategy for gold recovery from carbonaceous gold ores.

**Keywords:** carbonaceous gold ore; two-stage fluidized oxidation roasting; heat release characteristics; phase transformation; gold extraction

## 1. Introduction

Carbonaceous gold ore is defined as a kind of refractory gold ore because its carbonaceous matter can adsorb dissolved gold during extraction, resulting in the reduction of gold recovery [1,2]. With the depletion of easily leachable gold ore reserves worldwide, refractory gold ores are becoming increasingly significant to the mining industry. Approximately a third of gold production comes from refractory gold ore [3,4]. In addition, the mineralogy of gold deposits globally is becoming more complex and expensive to exploit [5]. Therefore, there is an urgent need to develop innovative and efficient gold recovery processes from refractory ores.

To eliminate the gold preg-robbing effect of carbonaceous matters in the gold leaching process, a lot of research has been done on the pretreatment methods of carbonaceous matters, such as oxidative roasting [3,4], biodegradation [6–8], and adding passivative or blanking agents [1,9]. Due to the strength of preg-robbing varying from ore to ore, these methods have their limitations, especially the long biological pretreatment cycle [10,11], the incomplete passivation of the passivator, and the possibility of hindering the dissolution of gold [12–14]. In contrast, oxidative roasting is a more reliable, mature, and adaptable technique for pretreating carbonaceous gold ores [15].

In the conventional rotary kiln for oxidation roasting, suitable roasting pretreatment conditions often need a long time for carbon and sulfur oxidation [15,16]. To accelerate the oxidation rate of carbon and sulfur, microwave roasting and oxygen-enriched air roasting



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**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). have been studied. In the study of utilizing microwaves in the pretreatment of carbonaceous sulphidic gold ores, microwave roasting had higher total carbon removal rates, faster heating rates, and lower specific energy consumption than conventional roasting [17,18]. However, Amankwah et al. [19] found that microwave heating was more difficult to control the temperature than conventional heating, resulting in local melting and sintering of the ore. This would increase the risk of the secondary encapsulation of gold particles. Similarly, oxygen-enriched air roasting is also an efficient decarburization and desulfurization technology for carbonaceous sulphidic gold ores. However, in an oxygen-enriched atmosphere, the combustion of carbon and sulfur in the refractory gold ore would release a large amount of heat, especially when the initial roasting temperature exceeded 600 °C and sintering of the ore was very likely to occur [20,21]. To solve this problem, Liu et al. [22] proposed a single oxygen-enriched roasting with the addition of Na<sub>2</sub>SO<sub>4</sub> to avoid sintering, and the formation of a porous structure was conducive to obtaining high extraction of gold (>95%) by alkaline Na<sub>2</sub>S leaching. Although the occurrence of sintering was effectively controlled by adding Na<sub>2</sub>SO<sub>4</sub> for industrial production, this may increase costs and complicate the production process.

It was noted that in the microwave and oxygen-enriched roasting, the material was in a static state, and the heat released by carbon and sulfur oxidation was accumulated because it could not be dissipated in time, resulting in a high local temperature and sintering in the material. Fluidized roasting may be a good choice to alleviate sintering to a certain extent. In fact, fluidized roasting has been widely used in iron ore reduction roasting [23–25], especially for refractory iron ore [26], iron tailings [27,28], and red mud [29]. Laboratory and semi-industrial tests of fluidized magnetization roasting have been carried out, which fully verified that fluidized roasting had a good mass transfer and heat transfer effect [30]. However, there were few reports on the fluidized oxidation roasting of carbonaceous gold ores, and the mechanism of the roasting process needs to be further explored. This is of great significance for the efficient decarburization of carbonaceous gold ores, thereby strengthening gold extraction.

In the present work, a two-stage fluidized oxidation roasting method was proposed for the pretreatment of carbonaceous gold ore. Consolidation of calcine is fatal to fluidized roasting. To avoid this problem, the pre-roasting (first stage) was conducted in a low oxygen environment (10% O<sub>2</sub>) to remove most carbonaceous matters from the raw ore, and then the pre-roasted products were roasted again to determine their heat release characteristics. The pre-roasted product (under the optimal pre-roasting conditions) was subjected to a secondary strengthened roasting (second stage), followed by leaching of gold extraction. The oxidation roasting mechanism and the phase transformation were investigated using X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and Brunauer–Emmett–Teller (BET) specific surface area measurement. Consequently, this study explores the feasibility of the two-stage fluidized oxidation roasting method for carbonaceous gold ore and provides a theoretical basis for the oxidation of carbonaceous matters during the two-stage roasting process.

### 2. Materials and Methods

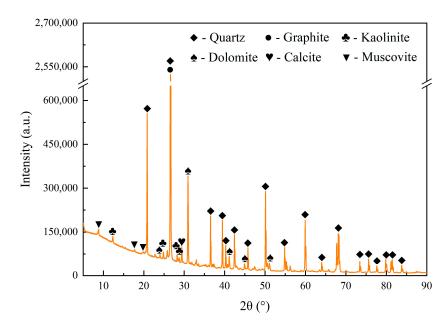
## 2.1. Materials

The carbonaceous gold ore used in this study was derived from a gold deposit in Shaanxi Province, China. The sample was ground to -0.074 mm with a gold content of 44%. Table 1 presents the chemical composition of the raw ore, which reveals Au, Ag, and SiO<sub>2</sub> constituting 4.0 g/t, 9.7 g/t, and 68.39%, respectively, with high content of C (7.02%) and S (1.60%). Combined with the analysis in Table 1 and Figure 1, the carbon in carbonate mainly existed in dolomite and calcite, and silicates mainly existed as muscovite and kaolinite in the raw ore. Table 2 shows that only 28.95% of the gold is exposed in the raw ore, while 49.88% and 18.00% of gold are encapsulated in silicates and carbonates, respectively. There is very little gold encapsulated in sulfides. The results of the carbon chemical phase analysis of the raw ore (Table 3) show that the carbon content in organic carbon, graphitic

carbon, and carbonate were 1.79%, 3.69%, and 1.23%, respectively. The literature [31] pointed out that in the cyanidation leaching of gold ore, when the organic carbon exceeds 0.2%, a serious gold preg-robbing phenomenon would occur. In addition, graphite carbon with a low graphitization degree could show similar adsorption performance to activated carbon, which would make gold in solution return to leaching residue again and reduce gold recovery.

Table 1. Chemica	l composition	analysis results	s of the raw ore.
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Components	Au <sup>a</sup>	Ag <sup>a</sup>	С	S	SiO <sub>2</sub>	CaO	MgO	$Al_2O_3$	Fe	Ba	К
Content (wt%)	4.0	9.7	7.02	1.60	68.39	5.67	2.83	2.32	1.22	1.11	0.49
Components	V	Р	Zn	Cu	Mn	Na	Hg	As	Sb	LOI	
Content (wt%)	0.23	0.22	0.034	0.017	0.014	< 0.05	0.0053	0.014	< 0.01	12.88	



<sup>a</sup>: g/t; LOI: Loss on ignition.

Figure 1. XRD pattern of the raw ore.

Table 2. Gold chemical-phase analysis results of the raw ore.

Phase	Exposed Gold	Encapsulated in Silicates	Encapsulated in Carbonates	Encapsulated in Sulfides	Total
Content (g/t)	1.19	2.05	0.74	0.13	4.11
Distribution (%)	28.95	49.88	18.00	3.16	100.00

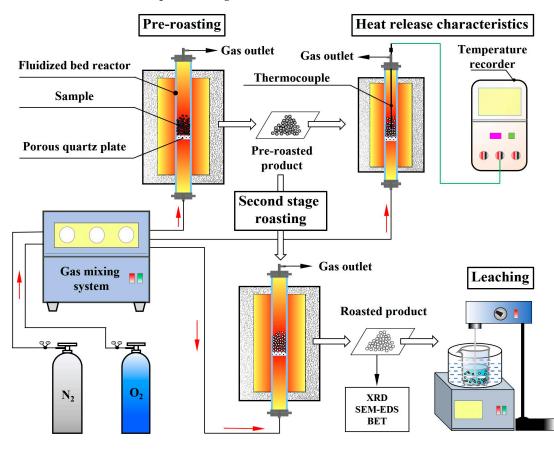
Table 3. Carbon chemical phase analysis results of the raw ore.

Phase	Organic Carbon	Graphitic Carbon	Carbonate Carbon	Total
Content (wt%)	1.79	3.69	1.23	6.71
Distribution (%)	26.68	54.99	18.33	100.00

The O<sub>2</sub> and N<sub>2</sub> used in oxidation roasting are all with a purity of 99.9%. The gold lixiviant purchased from Guangxi Senhe High Technology Co., Ltd. (Guangxi, China), is called "Jinchan" (JC) and is an environmentally friendly gold lixiviant [32].

# 2.2.1. Roasting Process

The experimental procedures are presented in Figure 2. Oxidation roasting included two stages: pre-roasting and second-stage roasting. Pre-roasting tests were performed in a vertical furnace (OTF-1200X-VT, KJ GROUP, HF-Kejing, Hefei, China) equipped with a fluidized bed reactor, the porous quartz plate in a fluidized bed reactor with a pore size of 10–15  $\mu$ m. Pre-roasting tests with a roasting time of 25 min, 30 min, 35 min, and 40 min were carried out. The other roasting conditions were fixed as follows: sample 100 g, temperature 650 °C, O<sub>2</sub> 10%, and total gas flow rate 2 L/min. The experimental operation of pre-roasting refers to the previous literature [33]. Then the heat release characteristics of pre-roasted products obtained at different roasting times were tested to determine the suitable pre-roasting conditions.



**Figure 2.** Schematic diagram of the fluidized oxidation roasting followed by leaching experimental system.

The tests for the heat release characteristics of the pre-roasted products were performed in a vertical furnace (OTF-1200X-S-VT, KJ GROUP, HF-Kejing, Hefei, China) equipped with a fluidized bed reactor, the porous quartz plate in fluidized bed reactor with a pore size of 10–15  $\mu$ m. The heat release characteristics of the pre-roasted products were characterized by directly measuring the temperature changes inside the material during the roasting process. The details of the experimental operation are presented as follows: first, 5.0 g pre-roasted product was added to the fluidized bed reactor, then a thermocouple equipped with a real-time temperature recorder (JK804, Changzhou Jinko Electronic Technology Co., Ltd. Changzhou, China) with an accuracy of  $\pm 0.1$  °C was inserted into the center of the material. The volume ratio of O<sub>2</sub> and N<sub>2</sub> was adjusted to 1:4, and the total gas flow rate was 0.4 L/min. After mixing evenly in the gas mixing system, the mixture was fed into the fluidized bed reactor. Next, the fluidized bed reactor was moved into the furnace with a preset temperature of 650  $^{\circ}$ C, and the internal temperature of the material was recorded in real time. The test was stopped after the reaction was carried out for 30 min.

By testing the heat release characteristics of the pre-roasted products, suitable preroasting conditions were obtained, and then the feed for the second-stage roasting was prepared under these conditions. The second-stage roasting was carried out for the sufficient decarburization of the pre-roasted product. The experimental steps of the second-stage roasting were the same as those of the pre-roasting, but the roasting conditions were different. In this section, the effects of roasting conditions, such as roasting temperature (600-800 °C), O<sub>2</sub> concentration (10-50 vol.%), and roasting time (5-25 min) on enhancing gold extraction, were investigated. The pre-roasted products were under suitable pre-roasting conditions, and the total gas flow rate was kept at 2 L/min.

### 2.2.2. Leaching Process

For the leaching experiment, the operation and parameters refer to the previous literature [33]. The leaching residue was dried at 105 °C and assayed for gold content, and the gold extraction efficiency was calculated using Equation (1).

$$\eta = \left(1 - \frac{\alpha}{\beta}\right) \times 100\% \tag{1}$$

where  $\eta$  is the gold extraction efficiency, %;  $\beta$  is the gold content in the roasted product, g/t, and  $\alpha$  is the gold content in leaching residue, g/t.

### 2.3. Analytical Methods

An infrared carbon sulfur analyzer was used to determine the total carbon and sulfur content of the samples. The chemical alterations in samples before and after roasting were investigated by thermogravimetry analysis (TG), derivative thermogravimetry analysis (DTG) and differential scanning calorimetry (DSC), and X-ray diffraction (XRD). The physical changes in the samples were determined by surface area and porosity analyzer (BET) and scanning electron microscopy (SEM).

### 3. Results and Discussion

#### 3.1. Roasting Experiments

In the one-step fluidized oxidation roasting test of carbonaceous gold ore [33], the highest gold leaching rate of 84.83% was obtained under the following conditions: roasting temperature of 650 °C,  $O_2$  concentration of 20%, roasting time of 75 min, and total gas flow rate of 2 L/min. Semi-industrial tests were conducted under these conditions, and it was found that calcines underwent consolidation, which was not present in the laboratory tests and seriously affected the normal operation of the fluidized roasting system. According to the literature [20], when the initial temperature exceeded 600 °C, consolidation in calcines was a common problem in the oxygen-enriched air roasting of refractory gold concentrates containing sulfur and carbon. It was inferred that the consolidation in calcines in the semi-industrial test was due to the more intense carbon and sulfur combustion under large feed conditions, which made it difficult to control the actual reaction temperature and resulted in the high local temperature of the calcines. Therefore, this study proposed using two-stage fluidized roasting to avoid the consolidation in calcines.

# 3.1.1. Analysis of Thermal Reaction Characteristics

The TG, DTG, and DSC cures of the raw ore under an air atmosphere are shown in Figure 3. Three obvious weight loss stages appeared on the DTG curve, located in the temperature ranges of 400–500 °C, 500–660 °C, and 660–800 °C. The weight loss stage in the range of 400–500 °C connected to the exothermic peaks on the DSC curve, which were caused by the exothermic oxidation of sulfides [34]. The DSC curve showed a strong exothermic peak in the range of 500–660 °C and connected to the second weight loss stage of DTG cure, indicating that a large amount of heat was released by the combustion of

carbonaceous matter. A significant weight loss was also observed in the DTG curve in the range of 660–800 °C, but the DSC curve showed that the reaction was endothermic, indicating that the response was caused by the decomposition of carbonates. The above results indicated that a large amount of heat was released when the carbonaceous matter in the raw ore was burned in the air.

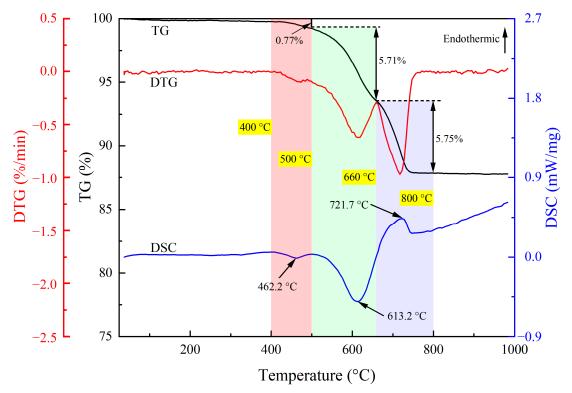
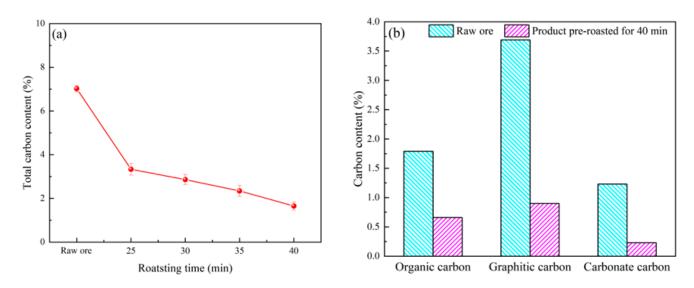


Figure 3. TG, DTG, and DSC cures of the raw ore under air atmosphere (10 K/min).

3.1.2. Pre-Roasting Experiments

Effect of Roasting Time on Decarburization

Some studies have shown that low-oxygen concentration could slow down the combustion rate of pulverized coal, promote uniform temperature distribution, and suppress the peak temperature [35]. Therefore, in the pre-roasting experiments, based on the optimal conditions of our previous study [33], the  $O_2$  concentration was fixed at 10% to slow down the combustion rate of carbonaceous matter and thus to avoid the consolidation in calcines caused by a large amount of heat release in a short time. In addition, according to the TG-DSC analysis of the raw ore, to maintain the stable combustion of carbonaceous matter and promote the decomposition of carbonates, the pre-roasting temperature was determined as 650 °C. Figure 4 shows the variation of the total carbon content for the pre-roasted product as a function of roasting time. The total carbon content gradually decreased with the increase in roasting time. When the roasting time was 40 min, the total carbon content decreased to 1.79%, and the carbon content reached a low level. If the pre-roasting time is further extended, the overall decarburization efficiency will be reduced. To ascertain the types of residual carbon in the pre-roasted products, the carbon chemical phase of the product pre-roasted for 40 min was analyzed, and the analysis results are shown in Figure 4b and Table 4. As shown, after pre-roasting for 40 min, the organic carbon, graphitic carbon, and carbonate carbon were greatly reduced, compared with the raw ore, but were still dominated by graphitic carbon, followed by organic carbon.



**Figure 4.** (a) The carbon content of the pre-roasted products and (b) carbon chemical-phase analysis results of the raw ore and the product pre-roasted for 40 min.

Table 4. Carbon chemical phase analysis results of the product pre-roasted for 40 min.

Phase	Organic Carbon	Graphitic Carbon	Carbonate Carbon	Total
Content (wt%)	0.66	0.90	0.23	1.79
Distribution (%)	36.87	50.28	12.85	100.00

## Heat Release Characteristics

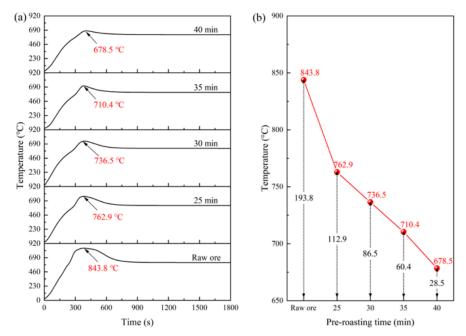
To select a pre-roasted product with suitable carbon content, the heat release characteristics of pre-roasted products with different roasting times were studied. Figure 5 shows the change process of the internal temperature of the material during the roasting of pre-roasted products with different carbon content. As shown in Figure 5, the combustion of carbon was very rapid, and the internal temperature of the material rose sharply in a short time. The internal peak temperature of the raw ore reached 843.8 °C, which was 193.8 °C higher than the preset roasting temperature and lasted for a long time above 650 °C. This indicated that the raw ore occurred in uncontrollable and violent combustion and released a large amount of heat in a short time once the temperature reached the ignition point of carbonaceous matter. Although this was beneficial in improving the decarburization efficiency, it was prone to cause the consolidation of calcine, thus increasing the risk of dissociated gold being secondary-encapsulated. With the decrease of carbon content in the pre-roasted products, the internal peak temperature of the material gradually decreased, indicating that reducing the carbon content in the material was conducive to controlling the actual reaction temperature inside the material. The internal peak temperature of the product pre-roasted for 40 min was  $678.5 \,^{\circ}$ C, which was the closest to the preset roasting temperature. To better control the actual reaction temperature of the material, avoid consolidation, and consider the overall roasting efficiency, the appropriate pre-roasting conditions were determined as: roasting temperature 650 °C, O<sub>2</sub> concentration 10%, and roasting time 40 min.

# 3.1.3. Second Stage Roasting Followed by Leaching Experiments

# Effect of Roasting Temperature

Figure 6 shows the effect of different roasting temperatures on gold extraction and total carbon content under an  $O_2$  concentration of 20% and a roasting time of 10 min. As shown, the gold extraction of the raw ore with high carbon content was only 4.55%, while the distribution of exposed gold in the raw ore was 28.95% (Table 2), which indicated that the carbonaceous matters have a strong gold preg-robbing effect. After roasting, the total carbon content decreased with the increase in roasting temperature, and the gold extraction

increased from 42.00% to 79.50% with the increase in temperature from 600 °C to 650 °C. However, the gold extraction decreased linearly when the roasting temperature exceeded 650 °C. This might be due to the closure of pores in the ore under high-temperature conditions, which hindered the reaction between the lixiviant and gold. Therefore, a roasting temperature of 650 °C was maintained in subsequent experiments.



**Figure 5.** (a) The internal temperature change process of materials during roasting, and (b) the comparison between the internal peak temperature of materials and the preset roasting temperature.

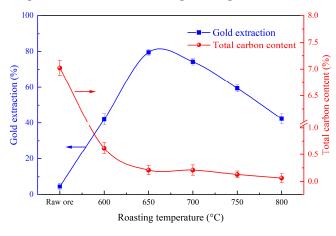


Figure 6. The effect of roasting temperature on gold extraction.

### Effect of O<sub>2</sub> Concentration

Figure 7 presents the effect of different  $O_2$  concentration on gold extraction and total carbon content under the conditions of a roasting temperature of 650 °C and a roasting time of 10 min. As seen in Figure 7, the total carbon content decreased with the increase in  $O_2$  concentration, and the gold extraction increased from 36.76% to 84.39% with the increase in  $O_2$  concentration from 10% to 25% and then decreased to about 78%–80% with the further increase in  $O_2$  concentration. In addition, after the  $O_2$  concentration exceeded 25%, the total carbon content of the roasted product did not change much, but the gold extraction decreased and did not improve further by extending the roasting time. When the  $O_2$  concentration was below 25%, it was possible to improve the gold extraction by prolonging the roasting time to remove the remaining carbonaceous matter, but it reduced the roasting efficiency. Thus, the  $O_2$  concentration of 25% was used in subsequent experiments.

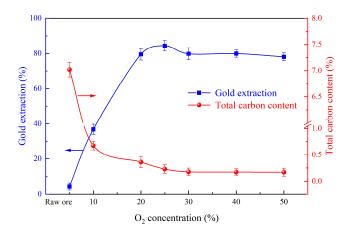


Figure 7. The effect of O<sub>2</sub> concentration on gold extraction.

### Effect of Roasting Time

The effect of different roasting times on gold extraction and total carbon content under the roasting temperature of 650 °C and O<sub>2</sub> concentration of 25%, as illustrated in Figure 8. The total carbon content decreased with the increase in roasting time, and the gold extraction increased from 42.44% to 86.10% with the increase in the roasting time from 5 min to 20 min, and then there was no significant change with further increase in the roasting time. From the above analysis of the results of the effect of O<sub>2</sub> concentration on gold extraction, it can be seen that when the roasting time was certain, the gold extraction decreased after an O<sub>2</sub> concentration exceeded 25%, and the roasting efficiency decreased if the gold extraction was increased by reducing the O<sub>2</sub> concentration. Considering the overall roasting efficiency, the second-stage roasting time of 20 min is reasonable. Therefore, the optimal parameters for second-stage roasting were determined as follows: roasting temperature 650 °C, O<sub>2</sub> concentration of 25%, and roasting time of 20 min.

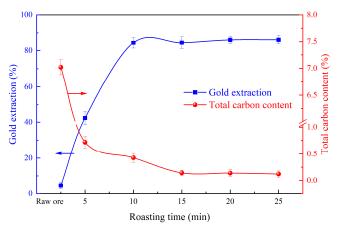


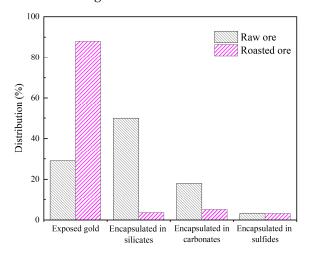
Figure 8. The effect of roasting time on gold extraction.

3.2. Mechanism Analysis of the Roasting Process

### 3.2.1. Gold Dissociation Analysis

Figure 9 and Table 5 show the analysis results of the gold chemical phase of the raw ore and the roasted product under optimal conditions. As shown in Figure 9, compared with the raw ore, the distribution of exposed gold in the roasted products was greatly increased by 59.16%, and the gold encapsulated in silicates and carbonates decreased from 49.79% to 3.64% and 18.16% to 5.14%, respectively. This indicates that under the optimal roasting conditions, the structure of silicates was destroyed, and most carbonates were decomposed, thus exposing the gold encapsulated in them. The distribution of exposed gold of roasted product under the optimal conditions agreed well with its gold extraction. It shows that the two-stage fluidized oxidation roasting of carbonaceous gold ore can not only efficiently

eliminate the gold preg-robbing effect of carbonaceous matter but also greatly improve the dissociation degree of gold, which is essential for improving the gold leaching efficiency of carbonaceous gold ore.



**Figure 9.** Gold chemical phase analysis results of the raw ore and the roasted product under optimal conditions.

Table 5. Gold chemical phase analysis results of the roasted product under optimal conditions.

Phase	Exposed Gold	Encapsulated in Silicates	Encapsulated in Carbonates	Encapsulated in Sulfides	Total
Content (g/t)	3.93	0.16	0.23	0.14	4.47
Distribution (%)	88.01	3.64	5.14	3.21	100.00

3.2.2. Phase Transformation Analysis

**Pre-Roasting Products** 

The phase transformation analysis of pre-roasting products is presented in Figure 10, and the main reactions that may be involved in the roasting process are Reactions (2) to (8). As seen in Figure 10, after roasting for 25 min, the diffraction peak intensity of dolomite decreased rapidly and disappeared when the roasting time increased to 40 min. When the roasting time was 25 and 30 min, the diffraction peak intensity of calcite was stronger than that of the raw ore, which was due to the decomposition of dolomite to generate part of calcite (Reaction (6)). After the increase in the roasting time, the intensity of the diffraction peaks of calcite gradually weakened, which was caused by the decomposition of some calcite (Reaction (7)). In addition, the diffraction peaks of anhydrite appeared in all pre-roasted products, indicating that the CaO produced by carbonates decomposition reacted with the SO<sub>x</sub> generated by the oxidation of sulfides (Reactions (4), (5), and (8)). Furthermore, the diffraction peak of kaolinite almost disappeared after 30 min of roasting, and the intensity of the diffraction peak of muscovite weakened with the prolongation of roasting time, which indicated that the structure of silicates had been destroyed to a certain extent after roasting.

$$C + O_2(g) = CO_2(g)$$
 (2)

$$2C + O_2(g) = 2CO(g)$$
 (3)

$$2FeS_2 + 5.5O_2 (g) = Fe_2O_3 + 4SO_2 (g)$$
(4)

$$3\text{FeS}_2 + 8\text{O}_2(g) = \text{Fe}_3\text{O}_4 + 6\text{SO}_2(g)$$
 (5)

$$CaMg(CO_3)_2 = CaCO_3 + MgO + CO_2 (g)$$
(6)

$$CaCO_3 = CaO + CO_2 (g)$$
<sup>(7)</sup>

$$2CaO + 2SO_2(g) + O_2(g) = 2CaSO_4$$
 (8)

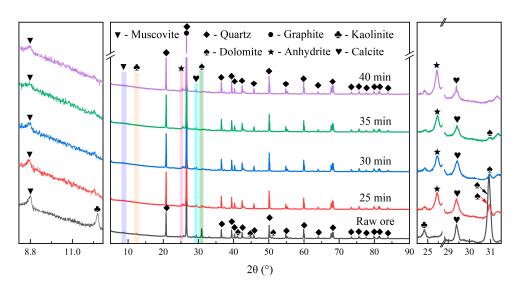


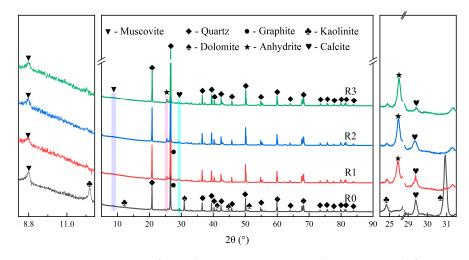
Figure 10. XRD patterns of the pre-roasting products.

## Second-Stage Roasting Products

The phase transformation of the roasted products in the second stage is shown in Figure 11. Here, the product roasted at 650  $^{\circ}$ C (R2) and the product roasted under optimal conditions (R3) were selected for comparative analysis with the raw ore (R0) and the product pre-roasted for 40 min (R1). It is obvious that the diffraction peaks of calcite still existed in the product roasted at 650 °C and in the product roasted under optimal conditions, but the intensity of calcite diffraction peaks in the product roasted under optimal conditions was very weak. This indicates that the decomposition of calcite was enhanced after the optimized roasting conditions. It can also be noted that the diffraction peaks of anhydrite were stronger in the product roasted at 650 °C and in the product roasted under optimal conditions, compared to the product pre-roasted for 40 min, indicating that the second-stage roasting promoted the sulfur fixation reaction of calcium-containing minerals. At the second stage, the roasting temperature of  $650 \,^{\circ}$ C and optimal roasting conditions, the diffraction peaks of muscovite and kaolinite in the roasted products weakened but did not disappear, compared with the product from the raw ore and the pre-roasted product for 40 min. Since the gold in the raw ore was mainly encapsulated in silicates and carbonates, it indicates that the secondary roasting intensified the decomposition or fracture of silicates and carbonates, which contributed to the exposure or dissociation of gold minerals.

### 3.2.3. Microstructure Analysis

According to the chemical analysis results of gold in the raw ore (Table 2), gold was mainly encapsulated in silicates and carbonates, so the morphological changes of silicates and carbonates before and after roasting were focused on. Figure 12 shows the SEM images and EDS results of the raw ore. From the element distribution of C, O, Si, Al, and K, it can be observed that carbonaceous matter and silicates were tightly embedded, which indicates that it is very difficult to separate carbonaceous matter from the raw ore by flotation or to weaken the gold preg-robbing effect of carbonaceous matter by adding passivative or blanking agent. In addition, it can also be seen from the element distribution of S and Fe that sulfides were closely embedded in carbonaceous matter and silicates, and the sulfur content in the carbonaceous matter was very high.



**Figure 11.** XRD patterns of samples: R0-raw ore, R1-product pre-roasted for 40 min, R2-product roasted at 650 °C, and R3-product roasted under optimal conditions.

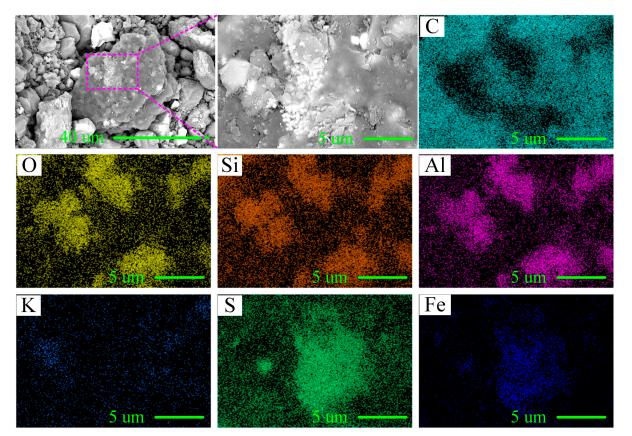
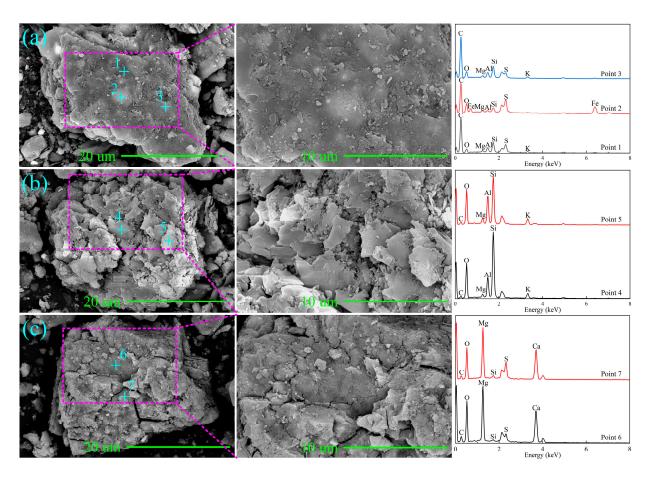


Figure 12. SEM images and EDS results of the raw ore.

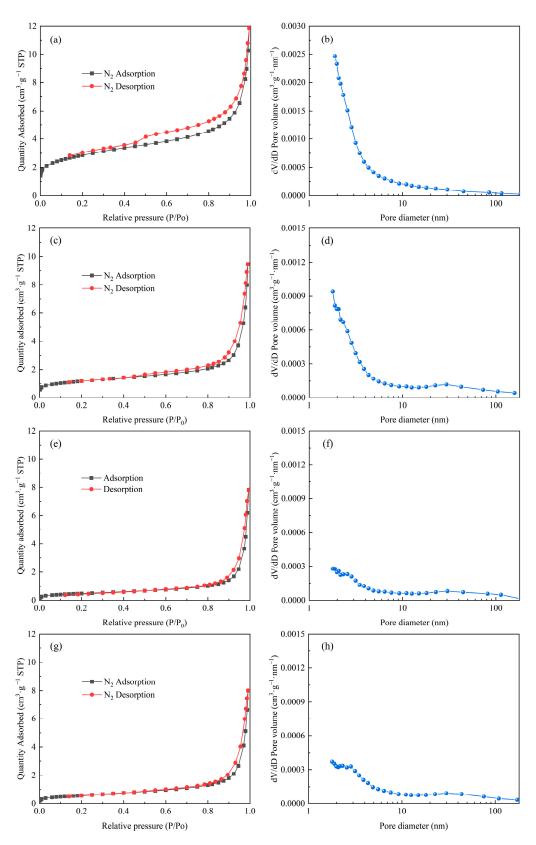
Figure 13 shows the SEM images and EDS results of the raw ore and product roasted under optimal conditions. As shown in Figure 13a, the carbonaceous matter and silicates in the raw ore were closely embedded and presented a relatively smooth surface. After roasting, the carbonaceous matter was burned out, and the silicates in the roasted product under optimal conditions (Figure 13b) showed a loose and porous structure. Figure 13c shows that the decomposition of dolomite produced many cracks and holes and captured a certain amount of sulfur. The loose and porous structure formed after roasting of silicates and carbonates increased the content of exposed gold, thus enhancing the gold leaching efficiency, which was consistent with the above gold extraction and gold chemical phase analysis results.



**Figure 13.** SEM images and EDS results of the raw ore (**a**) and the product roasted under optimal conditions (**b**,**c**).

## 3.2.4. BET and Porosity Analysis

The N<sub>2</sub> adsorption was carried out for the products roasting at different conditions to evaluate the relationship between gold extraction and the change of porosity. Figure 14 exhibits that the adsorption-desorption isotherms of the raw ore, the product pre-roasted for 40 min, the product roasted at 650 °C, and the product roasted under optimal conditions, all conformed to the reversible type IV isotherm, which is one of the main characteristics of mesoporous materials, and this can also be confirmed from their pore size distribution curves. Table 6 presents the BET-specific surface area and porosity parameters of products under different roasting conditions. As shown in Table 6, the BET-specific surface area of all samples decreased rapidly after roasting. This is due to the burning out of porous organic carbon and graphitic carbon in the ore [31]. It can be seen from Table 6 that the product pre-roasted for 40 min had a larger specific surface area, higher total pore volume, and a smaller average pore size than the products roasted at 650 °C and under optimal conditions, which was due to the residual organic and graphite carbon. As shown in Table 6, the product roasted under optimal conditions had approximately twice the specific surface area and smaller average pore diameter than those roasted at 650 °C, but the difference in total pore volume was not significant. This indicates that the product roasted under optimal conditions had more fine pores. These fine pores facilitated the entry of lixiviant into the ore and reacted with gold minerals.



**Figure 14.** N<sub>2</sub> adsorption–desorption isotherms and pore size distribution of samples: (**a**,**b**) raw ore, (**c**,**d**) product pre-roasted for 40 min, (**e**,**f**) product roasted at 650 °C, and (**g**,**h**) product roasted under optimal conditions.

Sample	Specific Surface Area (BET) (m <sup>2</sup> /g)	Total Pore Volume (cm <sup>3</sup> /g)	Average Pore Diameter (BJH) (nm)
Raw ore	9.94	0.0172	10.08
Product pre-roasted for 40 min	4.16	0.0145	16.76
Product roasted at 650 °C	1.17	0.0123	27.78
Product roasted under optimal conditions	2.13	0.0127	21.20

Table 6. The results of BET-specific surface area and porosity parameters of samples.

# 4. Conclusions

In this study, a two-stage fluidized oxidation roasting method followed by leaching was applied successfully to the gold extraction from carbonaceous gold ore. Reducing the carbon content in the ore by pre-roasting in a low-oxygen environment can effectively lower the peak temperature inside the material. The pre-roasting conditions were determined as: roasting temperature 650 °C,  $O_2$  concentration 10%, roasting time 40 min, and total gas flow rate 2 L/min. Through the single-factor experiments, the optimal second-stage roasting conditions were determined as follows: roasting temperature 650 °C,  $O_2$  concentration 25%, roasting time 20 min, and total gas flow rate 2 L/min. Under the optimal roasting conditions, the total carbon content decreased from 7.02% to 0.14%, and the gold extraction increased from 4.55% to 86.10%, indicating that the gold preg-robbing effect was effectively eliminated. In addition, the burning out of carbonaceous matter closely embedded with silicates and the decomposition of exposed gold from 28.95% to 88.01%, which created good conditions for gold leaching. Therefore, the two-stage fluidized oxidation roasting method is expected to be a new method to strengthen gold extraction from carbonaceous gold ores.

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