Study on dearsenification of gold concentrate by microwave roasting

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ABSTRACT. *Taking the gold concentrate containing arsenic in Hunan province as raw material, the mineral was roasted by microwave radiation to study the variation rule of arsenic and sulfur content in gold concentrate with temperature, power, holding time and other parameters, and the reaction mechanism of gold concentrate in microwave field heating was obtained by detecting and analyzing the products in the reaction process.*

Keywords: *Arsenic sulfide gold concentrate; Microwave roasting; Removing arsenic; Pyrolysis mechanism*

1. Introduction

Refractory gold ore refers to the ore with low gold leaching rate and high cyanide consumption when arsenic ore is extracted by conventional cyanidation method. With the development and utilization of a large number of gold resources, conventional gold ores that are easy to be prepared are becoming less and less, and the development and utilization of refractory gold ores containing arsenic has attracted extensive attention [1-5].

For the development and utilization of this kind of refractory arsenic-containing gold concentrate, it is mainly through the treatment of the raw ore before cyanide leaching, and the common pretreatment technology is mainly through physical, chemical and mechanical power methods to expose the gold wrapped in arsenopyrite or pyrite, so as to improve the gold leaching rate [6-8]. At present, the main pretreatment methods include pressure oxidation method, bacterial oxidation method, roasting oxidation method and chemical oxidation method [9-12]. However, these traditional treatment methods have high cost and low gold recovery. In this paper, the arsenic removal and sulfur treatment experiment of fine gold ores containing arsenic but difficult to be treated by microwave radiation is discussed, and the content is described as follows.

1.1 The mineral sample

The gold concentrate used in this study comes from a mineral sample after flotation in Hunan province. After drying, mixing, grinding and screening, the sample is prepared into a test sample. The results of multi-element analysis of the ore sample are shown in Table 1, the main elements of the sample include Fe, S and As. The mass percentage content was 19.94 %, 30.71 % and 8.87 %, respectively.

Table 1 Multi-element analysis of sample

The element	Au*	Ag*	As	S	Fe	Pb	Zn	Si	Ca	Mg
Content of	30 6	6.9	8.87	30.71	19.94	9.53	6.78	15.51	2.03	0.67

The band * has g/t and the other elements have %

1.2 Equipment and instruments

Microwave sintering equipment (KL-2D-6SJ, Guangzhou Kailing Microwave Equipment Factory); Tubular resistance furnace (SK-2-13, Shanghai East Star Co., LTD.); Standard vibrating screen (XSB-88, Shanghai Xinzheng Mechanical Instrument Manufacturing Co., LTD.); X-ray diffractometer (D/ Max-2550V, XRD analyzer of Japanese Physics); Gas flowmeter (Changzhou Thermal Meter General Plant); Scanning electron microscope (JSM-6700F, Seiko, Japan)

2. Test methods

2.1 Mineral detection and analysis

The phase analysis of the calcine and condensates was carried out with XRD detection conditions: Cu target and K ray were selected, the light source wavelength () was 0.154 nm, the incidence Angle was 2 ℃ ~ 80℃, and the detection step length was 0.03℃. The removal rate of arsenic and sulfur after microwave roasting can be calculated by measuring the content of arsenic and sulfur in minerals before and after roasting. The formula is as follows:

The band * has g/t and the other elements have %

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$$\eta = \frac{M \cdot \gamma_1 - m \cdot \gamma_2}{M \cdot \gamma_1} \times 100\%$$

In the equation, is the extraction rate of arsenic or sulfur. M represents the mineral mass of microwave roasting; M represents the quality of baking sand after microwave roasting; 1 represents the mass percentage content of arsenic or sulfur in minerals before roasting. Gamma 2 represents the percentage of arsenic or sulfur in microwave-treated calcines.

2.3 Microwave equipment and parameters

The operating parameters of microwave are: operating frequency (2450±50MHz); Input power (≤8kVA); Microwave output power (1000 ~ 6000KW); Single magnetron output power; (+ / - 5%) of 1000 w

Microwave schematic diagram

3. Experiment and discussion

3.1 Influence of microwave power on temperature rise

100 g gold concentrate was put into the sample tube for vacuum heating and the influence of different microwave power on the temperature rise of gold concentrate was shown in Figure 1:

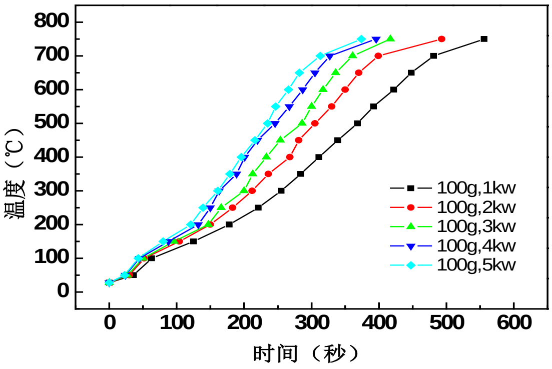


Figure. 1 Influence of microwave power on the heating behavior of gold concentrate

As can be seen from FIG. 1, under the same mass and atmosphere, the higher the microwave power, the higher the electric field intensity in the microwave cavity, the higher the energy density absorbed by the mineral per unit volume in the microwave field, and the higher the heating rate of the mineral. When the microwave power was 3 KW, the heating time of the mineral to 750°C was 6 minutes and 23 seconds, and the heating rate was 120℃/min. When the microwave power is 1 KW, the mineral temperature rises to 750℃ in 9 minutes and 26 seconds, and the temperature rise rate is 81℃/min. It can be seen that the mineral has a high wave absorption performance.

3.2 Influence of ore powder quality on temperature rise

1 KW microwave power, temperature rise end point 650℃, 4 sample dosage, namely 80 g, 120 g, 160 g, 200g and 240g, were selected for the test. The sample dosage was determined by the rate of temperature rise per unit mass. Three parallel tests were conducted, and the average value was taken as the test result, as shown in table 2

Table 1 Influence of mass on temperature rise

试验编号	质量（g）	微波功率（KW）	温度（℃）	通气量（L/min）	用时（秒）	单位质量升温速率 $(g^{-1}\cdot ℃/S)\times 10^{-3}$
NO1	80	1	650	4	631	12.48
NO2	120	1	650	4	416	12.62
NO3	160	1	650	4	334	11.79
NO4	200	1	650	4	285	11.05
NO5	240	1	650	4	244	10.76

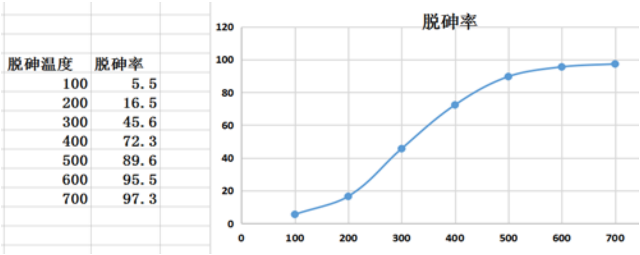
It can be seen from Table 2 that the average heating rate of minerals increases with the increase of mineral mass when the mineral samples are roasted under 1 KW microwave power. With the increase of mass, the larger the mineral volume and the same microwave power, the higher the absorbed power of the material. However, when the microwave power is fixed, the amount of ore increases and the microwave absorption fraction of ore powder per unit mass decreases, which leads to a decrease in the heating rate of ore powder. 1 KW microwave roasting power, N2 gas flow rate is 2 L/min, the end point of temperature rise is 650℃, unit mass rate of the fastest roasting sample volume is 120 g, the average rate of unit mass temperature rise is 14.55×10-3℃/S. Its heat loss is:

Q = a (tw - to) F,

In the formula, Q is heat dissipated, the unit is W, TW and to are surface temperature and ambient temperature of the heat dissipation surface respectively, the unit is ℃, F is the area of the heat dissipation surface, m^2, a is the comprehensive heat transfer coefficient, W /(℃×m^2)

3.3 Influence of heating endpoint on arsenic removal

Set the power of microwave at 1kW, the mass of roasted minerals at 120g, and hold the heat for 10 min. 250℃, 350℃, 450℃, 550℃, 650℃, and 750℃ were selected as the heating end points. The experimental results of arsenic and sulfur removal from mineral powder were shown in Figure 2.



脱砷温度	脱砷率
100	5.5
200	16.5
300	45.6
400	72.3
500	89.6
600	95.5
700	97.3

Figure. 2 Influence of temperature on arsenic removal

It can be seen from Figure 2 that the increase of the heating endpoint is conducive to the decomposition of minerals, and the removal rate of arsenic and sulfur in minerals increases with the increase of the heating endpoint. When the temperature is below 300℃, it is only 45.62%. At this temperature, the energy provided by microwave is not enough to decompose arsenic pyrite in a large amount, so the removal rate of arsenic is not high. When the temperature was from 400℃ to 500℃, the removal rate of arsenic increased significantly from 72.3% to 89.6%, and yellow smoke was observed in the settling bottle after the overcooling device during the test, as shown in Figure 3, indicating that pyrite and arsenopyrite in the temperature segment minerals were largely decomposed. When the temperature reaches 600℃, the arsenic removal rate in the mineral reaches about 97.3%. Meanwhile, metallic arsenic like silver mirror is attached to the wall of the test tube (as shown in Figure 4). With the increase of temperature, the sulfur removal rate increases slightly, and the sulfur removal rate is not high in the heating process.

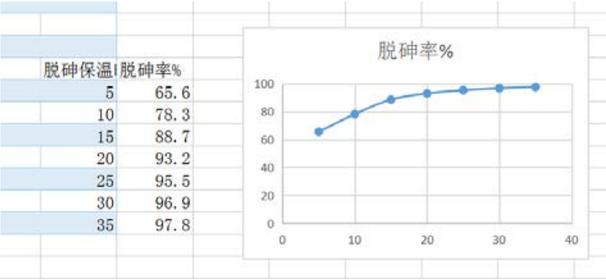
Figure. 3 Yellow smoke produced at 450℃

Figure. 4 Attachment produced at 650℃

It can be seen from Figure 3 and 4 that arsenic in minerals is mainly caused by thermal decomposition of minerals themselves induced by microwave, so increasing temperature has little influence on arsenic removal. Considering the economic reasons, the optimal arsenic removal temperature under the experimental conditions is 550℃.

3.4 Influence of holding time on arsenic removal and desulfurization

In the process of mineral arsenic removal by microwave roasting, besides the control of roasting temperature, the control of holding time should also be considered. In this experiment, when the microwave power was 1 KW, the roasting mass was 120 g, the heating point was 550℃, and the protective gas nitrogen flow was 4 L/min, the experiment was carried out for 4 min, 10min, 15min, and 30min. The results are shown in figure 5.



脱砷保温	脱砷率%
5	65. 6
10	78. 3
15	88. 7
20	93. 2
25	95. 5
30	96. 9
35	97. 8

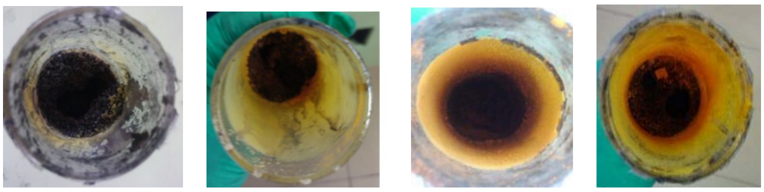
Figure. 5 Influence of holding time on arsenic and sulfur volatilization rate

FIG. 5 shows that the volatilization rate of arsenic in minerals increases with the extension of the holding time. When the heat preservation time reaches 15 min,

The removal rate of arsenic reached 95.2%, and increasing the heat preservation time had little influence on the removal of arsenic, so the heat preservation time was chosen to be 15 min.

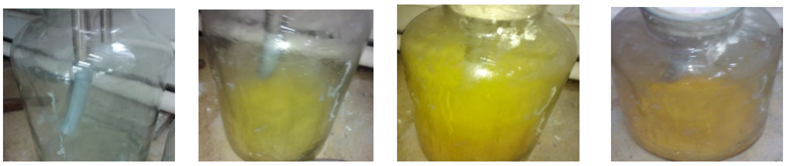
3.5 Decomposition mechanism of microwave arsenic removal

In the process of microwave roasting, the roasting products are different in different heating stages. The phenomena of substances on test tubes at different heating ends and collection bottles at different heating ends are shown in Figure 6 and 7. XRD tests are carried out on the baking sands and their products at different heating ends.



72/5000 A, 300℃ B, 400℃ C, 500℃ D, 600℃

Figure. 6 XRD tests were performed on the substances and products on the test tubes at different temperature ends.



A, 300℃ B, 400℃ C, 500℃ D, 600℃

Figure. 7 Phenomenon of collecting bottles at different temperature endpoints

Fig.10 XRD pattern of calcined sand at 400℃ Fig.11 XRD pattern of calcined sand at 600℃

It can be seen from FIG. 11 and 10 that there are different phase transitions for different heating endpoint minerals. Compared with the untreated mineral map, yellow sulfur elemental substance was produced in the roasting process when the temperature reached 400℃, indicating that the thermal decomposition process of pyrite was mainly at this temperature. When the temperature reached 600℃, the arsenopyrite and pyrite in the mineral were completely decomposed during 25 min roasting. At the same time, there were PbS and ZnS diffraction peaks in the whole heating process, indicating that no obvious sublimation or decomposition of lead sulfide and zinc sulfide occurred when the temperature was 600℃.

Test analysis to the experiment phenomenon, in the mineral pyrite decomposition temperature between 300 ℃ to 400 ℃, under the low oxygen environment, pyrrhotite and pyrite decomposition of sulfur, most of the sulfide in the form of elemental sulfur emerge from the mineral, a small amount of arsenopyrite under the condition of the decomposition of arsenic in the form of arsenic oxide isolated from arsenopyrite, a small amount of arsenic element and the elemental sulfur reaction generated arsenic sulfide compounds, removal from the mineral. When the temperature reaches 600℃, as the central temperature of the sample is higher than the outer temperature, the metal arsenic produced by the decomposition of arsenopyrite is partly condensed on the tube wall.

Based on the above analysis, it can be seen that the following reaction processes occur in the process of microwave protective roasting:

300℃-400℃, mainly the decomposition of pyrite: $N FeS_2 (s) = Fe_nS_{n+1} (s) + (n-1) s (s)$

$4 FeAsS (s) = 4FeS (s) + As_4 (s)$, $As_4+3O_2 (g) =2As_2O_3 (G)$

$As (s) + S (s) = AsS (s)$ or $2As (s) + 3S (s) = As_2S_3 (s)$

500℃ to 600℃, mainly arsenic sublimation: $As (s) = As (g)$, $As (g) +3O_2 (g) =2As_2O_3 (g)$

3.6 Influence of microwave roasting and conventional roasting on mineral appearance change

Influence of microwave roasting and conventional roasting on apparent change of minerals

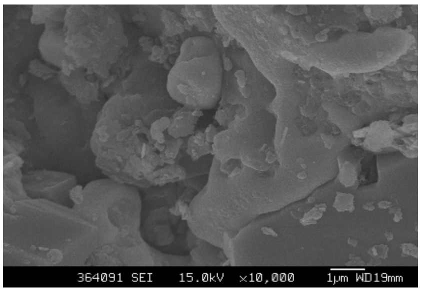


Figure. 8 SEM images of minerals after conventional roasting

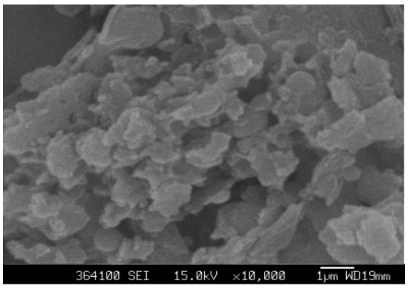


Figure. 9 SEM of baked sands after microwave irradiation roasting

FIG. 8 shows that the surface morphology of the ore samples roasted in the resistance furnace is smooth, with dense hooks and clear contours. Figure 9 shows that after the microwave radiation, a large number of pores, cracks and fragments appear on the surface of minerals.

Use the traditional roasting method such as resistance furnace roasting of gold concentrate, the temperature increase on mineral outside-in heat conduction, heat easily oxidized in the process of mineral surface, formation of low melting point metal oxide after oxidation at high temperature melting, with molten adhere to the mineral surface, to prevent the entry of oxidant, mineral internal reaction is difficult to fully, cause arsenic and sulfur elements in minerals cannot be completely removed.

Most sulfides in gold concentrate (FeS_2, FeS, PbS, ZnS, etc.) have relatively large dielectric constant. In the process of microwave radiation, significant temperature gradient is formed between minerals such as pyrite and ganglion, resulting in thermal stress on the interface between mineral phases and fractures, as shown in FIG. 13. The formation of cracks and pores increases the specific surface area of mineral reactions, making gaseous products such as arsenic in the roasting process easy to be separated from minerals.

4. Summary

1) The research on the heating behavior of gold concentrate in microwave roasting indicates that appropriate roasting quality and microwave power can promote the effective utilization of energy in the heating process of ore powder. From the analysis of temperature rise rate parameters per unit mass, the optimal power of microwave radiation with a mass of 120 g gold concentrate is 1KW.

2) In the dearsening test of gold concentrate by microwave radiation, the dearsening rate of calcined sand was 96.39%, respectively, after the temperature was kept at 550℃ for 15 min and the ventilation rate was 4 L/min.

3) After direct microwave radiation, XRD analysis shows that a large amount of pyrite is decomposed at 400℃. There are a lot of metal arsenic and arsenic oxide produced by roasting at 500℃.

4) According to the comparison test of conventional roasting, the absorption effect of sulfur-arsenic mineral in gold concentrate on microwave radiation is obvious, and its thermal decomposition reaction is low. SEM analysis and conditions of dearsenic-desulfurization show that the microwave radiation of gold concentrate heats up faster than conventional heating, which can improve the dearsenic-reaction rate of gold concentrate.

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