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Research status of carbonaceous matter in carbonaceous gold ores and bio-oxidation pretreatment

Hong-ying YANG[1], Qian LIU[1], Xiang-ling SONG[1], Jin-kui DONG[2]

1. School of Materials and Metallurgy, Northeastern University, Shenyang 110819, China;
2. Shandong Gold Mining (Laizhou) Company Limited, Laizhou, 261441, China

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Abstract: Carbonaceous gold mines are important refractory gold ores. The previous results demonstrate that the carbonaceous matter is mainly composed of elemental carbon, organic acid and hydrocarbons. The dissolved aurocyanide complex is robbed by adsorption of carbonaceous matter, which is similar to activated carbon in cyanide leaching of gold. The pretreatment methods of carbonaceous gold ores were introduced, including high temperature roasting, bio-oxidation, chemical oxidation, competitive adsorption, barrier inhibition and microwave roasting. Recently, bio-oxidation was developed rapidly due to its advantages such as mild conditions, simple processes, low energy consumption and friendly environment. The known microorganisms related with bio-oxidation pretreatment mainly are chemolithotroph bacteria such as *Thiobacillus ferrooxidans*, *Thiobacillus thiooxidans* and *Leptospirillum ferrooxidans*. The researches on decomposing and passivating carbonaceous matter were commenced by *Phanerochaete chrysosporium, Pseudomonadaceae* and *Streptomyces setonii.* Finally, the main problems were analyzed and the application prospect of this technique was looked forward.

Key words: carbonaceous gold ores; carbonaceous matter; pretreatment; bio-oxidization; research status

1 Introduction

The gold ores mainly are categorized into two types: non-refractory and refractory. With the increasing exhaustion of non-refractory gold ores, refractory gold ores have become main materials of the gold industry, and nearly one third of gold comes from refractory gold ores all over the world [1]. Carbonaceous refractory gold ores contain organic carbon and inorganic carbon from black (or carbonaceous) and sedimentary rock series [2]. The carbonaceous gold ores are refractory because dissolved aurocyanide complex is robbed by adsorption of carbonaceous matter, and this phenomenon is termed preg-robbing [3−5]. The famous carbonaceous refractory gold ores include the Carlin gold mine in Nevada of USA, the Kerr Anderson mine in Canada, the Ashanti and Prestea Gold Mines in Ghana and the Bakyrchik and Natalkinsk mines of the Soviet Union [6−8]. The plenty of large carbonaceous gold ores have been found in our country, which distribute mainly in the Yunnan−Guizhou−Guangxi, Shaanxi−Gansu−Sichuan, Qinling and central Hunan regions, as shown in Fig. 1 [2,9]. The gold reserves, carbon content and gold grade of international famous carbonaceous gold mines are listed in Table 1 [10−12]. How to develop and utilize carbonaceous gold ores economically and effectively has become an important problem facing the gold industry.

2 Composition and preg-robbing behavior of carbonaceous matter

The carbonaceous gold ores mainly exist in the form of composite minerals. The comprehensive analysis results of 168 Carlin type gold mines show that they have 8 element associations as shown in Table 2. The main gold- bearing minerals are carbonaceous matter in some fine- disseminated and metamorphosed rock type gold mines. For example, most of the gold of the Jerrit Cayon gold ore exists in carbonaceous matter as sub-

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Corresponding author: Hong-ying YANG; Tel: +86-24-83680373; E-mail: yanghy@smm.neu.edu.cn
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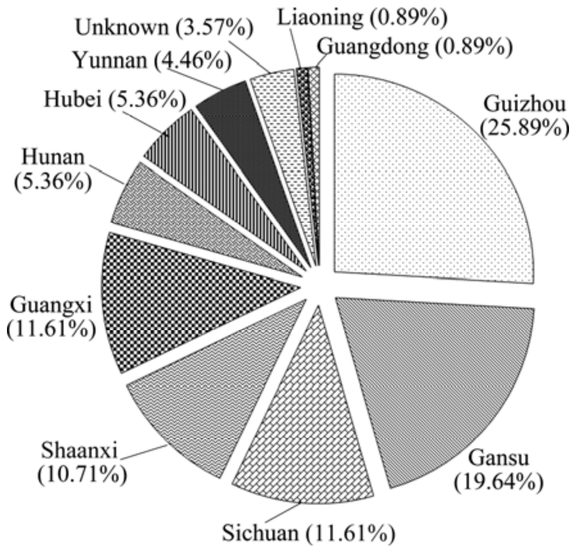

Fig. 1 Distribution of carbonaceous gold ores in China

Table 1 Gold reserves, carbon content and gold grade of international famous carbonaceous gold ores

Deposit name	Gold reserve/t	Carbon content/%	Gold grade/($g \cdot t^{-1}$)
Carlin gold mine in American	130.00	1.25−2.15	4.00−18.00
Kerr Anderson mine in Canada	17.23	0.05−7.99	28.35
Zimudang gold mine in Guizhou province, China	59.72	0.26−1.75	3.35−8.89
Getang gold ore in Guizhou province, China	23.67	1.01−6.80	1.14−27.34
Lannigou gold deposit in Guizhou province, China	22.48	0.06−1.13	3.78−10.70
Jinya mine in Guangxi province, China	18.05	1.54−2.33	1.55−7.16
Dongbeizhai gold mine in Sichuan province, China	7.21	2.80−2.96	1.90−5.00

Table 2 Element associations and examples of Carlin type gold mines

Element association	Example
Au−As−Hg−C	Lannigou gold deposit in Guizhou province, China
Au−Ag−As−Hg−C	Dongbeizhai gold mine in Sichuan province, China
Au−C	Yata and Getang mines in Guizhou province, China
Au−U−Hg−Sb−Mo−Y−Pt−(Os−Pd)−C	Laerma gold deposit in Gansu province, China
Au−Ag−As−Bi−Hg−C	Changkeng gold deposit in Guangdong province, China
Au−Hg−Ti−C	Zimudang gold mine in Guizhou province, China
Au−Sb−W−C	Longshan gold mine in Guangxi province, China
Au−As−Ag−Cu−Pb−Zn−Sb−C	Jinya mine in Guangxi province, China

microscopic particle; the gold contents of carbonaceous materials are 53.60 and 27.32 g/t in Banqi and Yata gold mines of China, respectively [2]. Recently, the researches are focused on the composition and properties of carbonaceous matter, while there are few studies on the mechanism of carbonaceous matter in cyanide leaching of gold, because of the fineness (0.002−2 μm), low content, crystal and amorphous state of carbon [13]. Apart from carbonates, carbonaceous matter of gold ores mainly includes elemental carbon, organic carbon, hydrocarbons and other biological substances, and the elemental carbon may be graphite or amorphous carbon. The most important carbonaceous preg-robbers are elemental carbon and organic carbon, and more than half organic carbon is humic acid. The preg-robbing behavior of elemental carbon is similar with activated carbon, while the humic acid forms complex with gold by functional groups [14−18]. Generally, if the content of organic carbon is above 0.2%, it will seriously disturb gold extraction cyanidation [2,9].

Carbonaceous matter has mainly two deleterious effects on gold extraction: the carbonaceous matter can lock up gold to inhibit leaching; the carbonaceous matter can absorb dissolved gold from precious solution [19,20]. The elemental carbon has been discovered in almost all carbonaceous gold ores, which are the natural activated carbon between anthracite and graphite [8,21]. ADAMs et al [22] reported that the preg-robbing capacity of some minerals was significantly increased because of the presence of activated carbon type carbonaceous matter. The preg-robbing capacity of carbonaceous materials is activated carbon firstly, humic acids secondly, while the hydrocarbons do not appear to interact with gold [23]. Another deleterious substance is humic acid which is the main component of humus. According to the solubility of humic acid in alkaline and acidic solutions, it is generally subdivided into three kinds: humus acid, fulvic acid and humin. The humus acid is black jelly, which dissolves in alkaline and weakly acidic solutions, but deposits below pH 3.0. Humus acid has strong adsorption capacity towards gold and silver because of its large surface area (337−340 m^2/g) [24,25]. The fulvic acid is soluble in aqueous solutions at all pH. WANG and GUAN [26] reported that fulvic acid could strongly dissolve and chelate gold at pH 3.0−8.4. The humin is an insoluble component in aqueous solutions at all pH. Humic acid can form stable chelate with gold by activated functional groups such as carboxyl, phenol-semi-quinone-quinone structures, phenolic and alcoholic hydroxyls, keto groups and aromatic rings [27]. The gold enrichment of humic acid is caused by the adsorption of humus acid and the migration and precipitation of fulvic acid [26]. The Infrared spectrum analyses indicated that hydrocarbons were mainly composed of long-chain alkanes. The hydrocarbons do not react with $Au(CN)_2^-$, and most of them coat the surface of activated carbon, which can decrease the preg-robbing activity [13,16]. The preg-robbing capacity of the carbonaceous matter generally increases with increasing maturity. The maturity of graphite carbon is similar to high rank anthracite, and the maturity of amorphous carbon is analogous to lower rank coals [14,16,28−30].

At present, the existing way of gold is still unclear in carbonaceous gold ores. Generally, the gold exists in the form of ultra-fine grade inclusions, adsorption or colloid adsorption. The preg-robbing mechanisms of carbonaceous matter, graphite and activated carbon are similar because they all possess aromatic nucleus and aliphatic side chains [2]. The adsorbing gold mechanism of activated carbon has not formed the unified viewpoint because activated carbon is affected by many factors such as ionic strength, pH and temperature in preg-robbing process; the analysis of activated carbon is difficult by traditional chemical techniques; and experimental conditions of different researchers have difference [31,32]. Currently, researchers have proposed the following mechanisms.

1) Ion pairs adsorption theory. Under conditions of high ionic strength, the aurocyanide complex is adsorbed in the form of the $M^{n+}[Au(CN)_2^-]_n$ [31,33]. This theory can reasonably explain the effect of cationic (as Na^+, K^+, Ca^{2+}, Mg^{2+}) on the adsorbing gold capacity of activated carbon [32].

2) Electrostatic adsorption theory. Under aerobic condition, the reaction (1) can occur in aqueous suspension of activated carbon. The activated carbon has positive charge because of the electron transfer, which can promote the attraction on $Au(CN)_2^-$ with negative charge [34,35].

3) Ion exchange theory. Chemical bonds are formed by interaction between functional groups of activated carbon and aurocyanide complex. For example, the $Au(CN)_2^-$ is adsorbed onto the activated carbon by ion-exchange with hydroxyl of chromene active center [36]. All the samples with graphite structure have a certain preg-robbing activity, and the chemical bonds related to graphite structure play an important role, which may be related to bonding electrons [37].

$$O_2+2H_2O+2e=H_2O_2+2OH^- \quad (1)$$

3 Pretreatment methods of carbonaceous gold ores

In order to eliminate or passivate carbonaceous matter, many pretreatment methods are adopted, which are divided into two types: 1) removing or decomposing carbonaceous matter in ores; 2) passivating carbonaceous matter partly or completely [2]. The passivation cannot destroy carbonaceous matter, so inclusion gold is not liberated. At present, several methods have been used to pre-treat carbonaceous matter such as high temperature roasting, bio-oxidation, chemical oxidation, competitive adsorption, barrier inhibition and microwave roasting.

High temperature roasting is a reliable technique to decompose carbonaceous matter and improve the leaching efficiency of gold for mature process and strong adaptability. However, this method also has following defects: 1) the energy consumption is high, while some reaction products pollute environment; 2) excessive roasting easily forms physical encapsulation and crack closure of pores which can lead to the second refractory of gold; 3) the ashing conditions of carbonaceous matter are very harsh, if conditions are improperly controlled, it will improve the activity of carbonaceous matter and decrease the leaching rate of gold further [13,38]; 4) the roasting temperature of gold ores cannot exceed 700 °C, otherwise the gold will form arsenic-gold alloy and volatilize easily. The research showed that the ideal calcination temperature was 360 °C [39].

Bio-oxidation method is a new process of pre-treating carbonaceous refractory gold ores. This method oxidizes and passivates carbonaceous matter to reduce the adsorbing gold capacity by microbial metabolism. Compared with other methods, the process has many advantages such as mild conditions, simple process, low energy consumption and friendly environment [13,40]. Therefore, the application of bio-oxidation method can solve the restriction of resource−energy−environment triangle in mineral processing industry effectively [41].

Chemical oxidation method can oxidize organic carbon and passivate graphite type carbon. The chlorination oxidation method uses Cl_2 or HClO to form thin layer of chloride−carbon complex on the surface of carbonaceous matter by oxidizing preg-robbing functional groups and replacing sulfur of the organic carbon, thus realizes the passivation of carbonaceous materials [13,42,43]. The high temperature and high-pressure oxidation reduction method employs nitric acid to oxidize and/or passivate the carbonaceous materials. This method can partially deactivate carbon, but it is able to activate carbonaceous matter in some cases.

The competitive adsorption method uses competition effect to decrease the preg-robbing capacity of carbonaceous matter by adding adsorbents such as activated carbon and ion exchange resin which have stronger adsorption capacity than carbonaceous matter in the cyaniding process. The ion exchange resin is more effective than activated carbon in respect of reducing or removing harmful effect of carbonaceous matter. In the carbon-in-leach process, activated carbon is used to compete with carbonaceous matter for gold extraction, but it has less effect on highly preg-robbing ores with high concentration of micro-fine particle carbon [4,17].

The preg-robbing capacity of carbonaceous matter can be passivated by adding blanking agents such as kerosene, diesel and some of aromatic hydrocarbon derivatives which adsorb selectively and coat onto the surface of carbonaceous matter, and this method is referred to as barrier inhibition process. The organic substances can inhibit the preg-robbing activity of carbonaceous matter by forming electrostatic repulsion or adsorbing preferentially on the surface activity sites [2,8,44]. In addition, the inhibitors with strong hydroscopicity are chosen to reduce or remove the preg-robbing of carbon in gold ores because of the hydrophobicity of element carbon surface [45]. The solid−liquid separation is very difficult because the fuel oil is lighter than water after leaching.

Microwave roasting can directly heat materials by energy dissipation of microwave in the materials. The results showed that the treatment effect of samples is related to the microwave power, processing time and sample characteristic. Carbon is known as very good microwave absorbers, so it can be heated and oxidized rapidly [24,44]. The method has many advantages such as fast decarburization speed, high heating rate and low energy consumption compared with the conventional roasting [46]. Currently, this method still has several disadvantages in terms of study and development of equipment, which restricts its industrialization process.

4 Research status of bio-oxidization pretreatment on carbonaceous gold ores

4.1 Species and source of microorganisms

The leaching microorganisms can be divided into inorganic autotrophy and inorganic heterotroph according to the nutrition type. At present, the known microorganisms related to bio-oxidation pretreatment include mainly *Thiobacillus ferrooxidans*, *Thiobacillus thiooxidans*, *Leptospirillum ferrooxidans*, *Sulfolobus acidocaldarius* of *Sulfolobus* and *Sulfobacillus* sp., etc., and most of these strains are chemolithotroph bacteria [19]. *Thiobacillus ferrooxidans* and *Thiobacillus thiooxidans* are very similar in morphology, physiology and biochemistry characteristics, so both bacteria are called *Thiobacillus ferrooxidans*. In recent years, some researches have indicated that several heterotrophic bacteria, actinomycetes and fungi such as *Phanerochaete chrysosporium*, *Pseudomonadaceae*, *Streptomyces setonii* could also passivate or decompose carbonaceous matter. Most of the strains are natural or artificial domestication in bio-oxidation pretreatment. However, there is less research on high efficient engineering bacteria which are constructed by biotechnologies such as cell engineering, gene engineering and protein engineering. Natural strains have strong adaptability without long-term domestication. As for domestication strains, the domestication target is the key problem [47]. The conventional microbes have many defects such as slow reaction rate, poor heat resistance and susceptiblity to heavy metal ions disturbances which are solved by breeding of high efficient engineering bacteria with strong adaptability and high oxidation activity. Excellent arsenic-resistant strains have been bred by ultra violet, laser and microwave [48].

4.2 Mechanism

At present, the mineral chemotrophic bacteria such as *Thiobacillus ferrooxidans*, *Thiobacillus thiooxidans*, *Leptospirillum ferrooxidans* cannot efficiently oxidize carbonaceous components, and their mechanism may be passiviation. The mechanism of fungi on the carbonaceous matter includes mainly passivation, alkalization, chelation and oxidation of biological enzyme-free radicals.

4.2.1 Passivation mechanism

The gold adsorption of elemental carbons mainly depends on the activity sites related to adsorption because its maturity is similar to the anthracite [20]. The thallus and metabolites such as extracellular polymeric substances (EPS) which can adsorb and coat the surface of carbonaceous matter to reduce the preg-robbing capacity by changing physicochemical properties of minerals [49−53]. The passivation mechanisms of these organic matters have two following types: 1) the preg-robbing capacity of carbonaceous matter was inhibited by electrostatic repulsion with organic matter [2]; 2) the organic matter could reduce the adsorbing gold capacity of carbonaceous matter by competitive adsorption [8].

4.2.2 Oxidation of biological enzyme-free radicals mechanism

Some fungi can synthesize and secrete many biological enzymes which consist of various degradation systems such as lignin-degrading enzyme system and aromatic-ring-degrading enzyme system. The interaction process between enzymes and substrates produces massive free radicals, which can destroy the structure of carbonaceous matter to reduce the preg-robbing capacity. And the specific mechanism is as follows: Firstly, bio-enzyme—$P(Fe^{3+})$ reacts with H_2O_2 to lose two electrons (2e) and form cation free radical —$P(O{=}Fe^{4+}\cdot)$. $P(O{=}Fe^{4+}\cdot)$, subsequently reacts with a substrate molecule RH, which results in electron transfer from RH to $P(O{=}Fe^{4+}\cdot)$, thus free radicals R· and $P(O{=}Fe^{4+})$ forms. Finally, $P(O{=}Fe^{4+})$ returns to the initial resting state by reaction with another molecule RH [19,20,54−56].

4.2.3 Alkalization and chelation mechanisms

Fungi, single cell bacteria and *Streptomyces* sp. can produce and secrete nitrogenous bases such as polypeptide and polyamine which are involved in the degradation of carbonaceous matter by improving the pH of the reaction system. Some of microorganisms can produce chelates in metabolic process. And these chelates can chelate and eliminate multi-valence metal ions which play the role of bridge in carbonaceous matter, thus achieve destroy structure of carbonaceous matter and reduce the preg-robbing capacity.

Recently, the authors studied the degradation of *Phanerochaete chrysosporium* on activated carbon. The results indicated that the degradation rate of activated carbon reached 27.59% after 14 d incubation with *Phanerochaete chrysosporium*. It can be seen that the micro-crystalline structure and species and contents of functional groups of activated carbon were changed by XRD and FTIR analyses. The study was also found that the pH of culture medium increased from 4 to 8−9, which indicated that some of the alkaline materials were produced. Therefore, the pretreatment of *Phanerochaete chrysosporium* on carbonaceous matter may be the result of combined effect of passivation, alkalization and oxidation of biological enzyme-free radicals.

4.3 Research status at home and abroad

BRIERLEY and KULPA [51] found that most of heterotrophic bacteria from the *Pseudomonas* family could coexist with the gold ores, and these bacteria were able to deactivate the active sites on carbonaceous matter. They used *Thiobacillus ferrooxidans* to oxidize sulfides and the gold extraction rate increased from 0 to 55.5% for the untreated control. Afterwards, the carbonaceous matter was deactivated using microbial consortium which is comprised of *Pseudomonas maltophilia*, *Pseudomonas oryzihabitans*, *Achromobacter* species and *Arthrobacter* species, and the leaching rate of gold was improved from 55.50% to 74.40%. YEN et al [53] pre-treated refractory and double refractory ores using *Trametes versicolor* agent and/or culture media. Firstly, *Trametes versicolor* culture media (with the fungal agent) were used to deactivate the preg-robbing carbonaceous components, and the gold extraction rate was between 54.10% and 64.50%. Secondly, the refractory sulfides of the ores were decomposed by *Trametes versicolor* culture media (without the fungal agent). Greater gold extraction occurred when the ore samples were treated by a combination of bio-treatment and bio-oxidation with *Trametes versicolor*, resulting in 87.00%−95.25% extraction. The gold leaching rate of carbon-bearing high-arsenic refractory gold concentrate of Guangdong province is only 15.02%. YANG et al [52] employed HYK−2 flora to treat this refractory gold ores, and the bacteria could excrete a mass of organic substances when the metal sulfides were oxidized. The bacteria and colloidal phase culture medium could attach to the surface of organic carbon and passivate it, and the leaching rate of gold reached 94.41%. The Dongbeizhai gold mines are typical double refractory ores, and the gold leaching rate is almost 0. This gold ore has high concentration of harmful elements such as arsenic, sulfur and carbon, and the gold is present as submicroscopic colloidal gold and native gold in the pyrite. WANG et al [49] found that the content of organic carbon was nearly no change before and after bacterium treatment, but the recovery rate of gold was more than 95%. This indicated that *Thiobacillus ferrooxidans* had passivation on organic carbon. Conversely, AMANKWAH et al [19] held that *Thiobacillus ferrooxidans* could not oxidize and deactivate carbonaceous matter effectively. It still was preg-robber during cyanide leaching process.

AMANKWAH et al [19] used two-stage bio-oxidation to treat double refractory gold concentrates which contain 65.30 g/t Au, 6.10% C and 11.90% S, and the main sulfide minerals are pyrite and arsenopyrite. In the first stage, chemolithotrophic bacteria were used to oxidize sulfides, and cyanidation resulted in 81.1% gold extraction. The action of *Streptomyces setonii* caused reduction of carbonaceous matter content in the second stage. The combined effect of two steps resulted in an overall gold recovery of 94.7% after cyanidation. AMANKWAH and YEN [57] employed *Streptomyces setonii* to pre-treat lignite, bituminous and anthracite. They found that carbon dioxide was produced in the process of degradation, and the degradation rates of lignite and bituminous were higher than anthracite. SARPONG et al [20] used lignite, sub-bituminous, bituminous and anthracite as substitute to study the influence of *Phanerochaete chrysosporium* on the preg-robbing capacity of carbonaceous matter. The results indicated that *Phanerochaete chrysosporium* could decrease the preg-robbing capacity by about 90%. The *Phanerochaete chrysosporium* could secrete enzymes to degrade gold-bearing wood chips, which increased contact between gold and cyanide solution. Although the interaction between chips and *Phanerochaete chrysosporium* caused relatively high mass losses of chips, but the leaching rate of gold could only be improved by 10% [58].

5 Prospect of bio-oxidization pretreatment technology

Compared with the traditional process, bio-oxidation pretreatment technology has obvious advantages in pre-treating carbonaceous gold ores, thus this method is popularized and utilized rapidly in the world and has broad prospects for development. The possible development direction of bio-oxidation pretreatment process in the future mainly includes the following aspects: 1) It is necessary to study the mechanism and oxidation kinetics of bio-oxidizing carbonaceous materials, the future potential trends of dynamic researches are building energy consumption model from biological metabolites [59,60]; 2) Construction of high efficient engineering bacteria with fast growth, strong adaptability and high oxidability is an important development direction by various biotechnologies such as genetic-engineering, gene-recombination and mutation; 3) At present, the microbial reactors have many defects such as low mass transfer coefficient, high shear stress and low amplification accuracy, so the new microbial reactors with high mass transfer rate, low shearing force, low residence time and low power consumption will become a promising development direction in future. With the development of bio-oxidation technology, it will inevitably become an effective way to develop and utilize refractory gold resources, and bring good economic and social benefits for gold industry.

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碳质金矿的碳质物及生物氧化预处理研究现状

杨洪英[1]，刘 倩[1]，宋襄翎[1]， 董金奎[2]

1. 东北大学 材料与冶金学院，沈阳 110819；2. 山东黄金矿业(莱州)有限公司，莱州 261441

摘 要：碳质金矿是一种重要的难处理金矿。研究发现，其碳质物主要包括元素碳、有机酸和烃类物质。在氰化浸金过程中碳质物可通过类活性炭的吸附方式将已溶解的金劫走。目前，已有的预处理方法主要有高温焙烧法、生物氧化法、化学氧化法、竞争吸附法、覆盖抑制法、微波加热法。生物氧化法因具有条件温和、流程简单、能耗低、环境友好等优点得以迅速发展。与生物氧化预处理有关的微生物主要有氧化亚铁硫杆菌、氧化硫硫杆菌、氧化亚铁钩端螺旋菌等化能无机自养菌。有关黄孢原毛平革菌、假单胞菌、多毛链霉菌在碳质物降解和钝化方面的研究也已展开。最后，分析了该技术存在的问题，并对其应用前景进行了展望。

关键词：碳质金矿；碳质物；预处理；生物氧化；研究现状

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