

Bio-mineral processing of carbonaceous refractory gold ore applying fungal enzymes

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(真菌放出型酵素反応を導入した炭素質金鉱石のバイオミネラルプロセッシング)

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論 文 内 容 の 要 旨

The bio-treatment of carbonaceous matter in double refractory gold ores (DRGO) is one of the most environmentally safe beneficiation routes for improving gold recovery. As a result, many bacterial and fungal species have been studied for their potential application to the gold ore on an industrial scale. A white-rot fungus, *Phanerochaete chrysosporium*, is one of these microbes, and its carbonaceous matter decomposition ability is due to the secretion of extracellular lignin-degrading enzymes. While fungal enzymes have been applied to water treatment in the pulp industry, it has never been successfully utilized for solid substrates in any industry. Several attempts have been made to apply this fungus to the treatment of DRGO in the past 3 decades. In most cases when the real ore was used, the characterization of the effectiveness of the fungus was based only on the gold recovery rather than direct analysis of the carbonaceous matter transformation. Therefore, to improve the understanding of the carbonaceous matter transformation, the cell-free spent medium (CFSM) of *P. chrysosporium* was used to treat DRGO sequentially, and the alteration in the carbonaceous matter is elucidated in this thesis.

It was important to determine the effectiveness of the CFSM as an oxidant for the carbonaceous matter in the DRGO. As such, powdered activated carbon (PAC) was used as a surrogate due to its graphitic structure during the CFSM treatment in **Chapter 3**. It was found that the CFSM treatment decomposed the aromatic C=C bonds in the PAC confirmed by solid ^{13}C -nuclear magnetic resonance (^{13}C -NMR), which resulted in decreases in the specific surface area and formation of large micron-sized pores on the bio-treated PAC. Additionally, some of the bio-molecules, secreted by the fungus into the CFSM, passivated the surface of the treated PAC and thus increased the surface negativity. Due to these factors, the gold uptake decreased significantly, implying that the CFSM treatment degraded the aromatic carbon as a solid substrate.

In **Chapter 4**, the sequential bio-treatment of the DRGO by sulfide oxidation using a thermophilic archaeon followed by enzymatic degradation of the carbonaceous matter was investigated using a combination of water chemistry, quantitative evaluation of minerals by scanning electron microscopy (QEMSCAN), thermogravimetric-differential thermal analysis (TG-DTA), scanning electron microscopy (SEM), DNA extraction and cyanidation. When the DRGO was pre-treated by bio-oxidation of sulfides with *Acidianus brierleyi* (DA) and only CFSM (DC), the gold recovery was improved from 24% to 77% and 38%, respectively. This was due to the inability of the CFSM to effectively breakdown the sulfides, which were the major gold-bearing minerals. When sulfide oxidation using *A. brierleyi* was followed by carbonaceous matter degradation using the CFSM (DAC) and vice versa (DCA), the gold recovery was 76% and 45%,

respectively. The DCA sequence was judged to have been unsuccessful. The DAC sequence, on the other hand, showed that the CFMSM treatment had converted the carbonaceous matter into an alkaline soluble substance which could be removed by washing. After 1 M NaOH washing, the gold recovery for DAC was 92% while 60% was observed for DCA. This result indicates that the sequence utilizing the iron-oxidizing microbe before the CFMSM was the ideal way to pre-treat the DRGO.

The transformation of the carbonaceous matter during the DAC sequence was investigated using QEMSCAN analysis, Raman spectroscopy and three-dimensional fluorescence spectroscopy in **Chapter 5**. The results show that the carbonaceous matter was initially hosted in illite and the amount of carbon relative to the amount of illite affected the texture of the carbonaceous illite. The enzymatic decomposition of the carbonaceous matter was found to be accelerated in the DRGO if the sample had undergone a prior treatment to decompose Fe sulfides and arsenopyrite. This is because the enzymes are susceptible to arsenic poisoning and therefore, the oxidative dissolution of arsenopyrite aided the enzymatic reaction. The lignin-degrading enzymes preferentially attacked the defects in the graphitic structure of carbonaceous matter and in the process, produced humic-like substances. The humic-like substances that are produced by the CFMSM treatment acted as one of the binding agents in the agglomeration of the carbonaceous aluminosilicate residue (C-Si-Al). This new C-Si-Al appeared to be the main product of the carbonaceous illite decomposition, and its retention of the humic substances explains why the alkaline washing step was needed to improve gold recovery from DAC from 76% to 92%.

In **Chapter 6**, a preliminary investigation was conducted to determine if enzymatic treatment would be viable for carbonaceous base metal ores like copper or nickel sulfides. Such a beneficiation route would involve having to decompose the carbonaceous matter before the copper or nickel recovery. This means that the DCA sequence would have to be considered and in **Chapter 4**, it was found to be ineffective due to the possible inhibition of *A. brierleyi*. Therefore, it was necessary to determine the leading cause of *A. brierleyi* inhibition after the CFMSM treatment. The three main impediments that were identified after the CFMSM only treatment were humic-like substances from the decomposition of the carbonaceous illite, biomolecules like carbohydrates and organic acids which aided in the formation of biofilms and large agglomerates, and finally, the fungal biomass. Washing the CFMSM-treated solids with 1 M HCl and 1 M NaOH removed biofilms, broke down the aggregates and extracted the humic acids, but sulfide oxidation by *A. brierleyi* was still impeded. Therefore, it was concluded that the fungal biomass itself was the main impediments, and its negative effect became prominent if it made up more than 1wt% of the ore. Based on this information, the application of the *A. brierleyi* to the CFMSM-treated carbonaceous base metal ore would not be successful without first reducing the amount of fungal biomass significantly.

In the **Chapter 7**, this work is summarized.