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学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称 博士（工学） 氏名 ZOLETA Joshua Bravo

学 位 論 文 題 名

Selective Cementation of Gold Using an Iron Oxide and Zero-Valent Aluminum Galvanic System from Gold-Copper Ammoniacal Thiosulfate Solutions

(酸化鉄と0価アルミニウムを用いた金-銅含有アンモニウムチオ硫酸溶液からの金の選択還元析出)

In conventional gold ore processing, two critical problems need to be addressed: one is the use of toxic chemicals during the extraction process, and the other is the depletion of gold ore reserves that can be quickly processed. Thus, the development of eco-friendly alternatives to extract gold from refractory ores is needed. Many studies have been conducted to find alternative solvents, such as thiourea, thiocyanate, halogen-based and thiosulfate. Among the alternatives, ammonium thiosulfate has been highlighted because it has been shown to have higher selectivity and leaching rate to gold, and most importantly, it is less corrosive and non-toxic to humans. While methods to recover gold from the leachate have yet to be established, this causes limitations to commercial operations.

A recent study reported that gold recovery could be achieved by cementing the gold ions via galvanic interactions between activated carbon (electron mediator) and zero-valent aluminum (electron donor) in the gold-copper ammoniacal thiosulfate system. The research also highlighted that metal ions can be recovered via galvanic interactions using Al and other conductive materials.

Considering the semiconductive properties of hematite and magnetite, it may be possible to use them as electron mediators. This dissertation evaluated the technical feasibility of two iron oxides (hematite and magnetite) as electron mediators for the enhanced cementation of gold from ammonia thiosulfate leachate using zero-valent aluminum as an electron donor.

Chapter 1 describes the problem statement and this study's objective. Literature on the recent studies of gold leaching and recovery from leachate using ammonium thiosulfate solutions was reviewed.

In Chapter 2, cementation experiments were conducted using iron-oxides as electron mediators and aluminum as electron donors to investigate metal recovery from a model ammonium thiosulfate leachate containing gold and copper ions. Solution analysis and SEM-EDX observation for cemented material confirmed that copper and gold were cemented from the solution in a reference experiment using activated carbon as an electron mediator. On the other hand, when iron-oxides like magnetite and hematite were used as electron mediators, copper deposition was limited, and gold was selectively deposited. This indicates that selective recovery of gold is possible by using iron-oxides and aluminum.

In Chapter 3, electrochemical experiments were conducted to evaluate the galvanic interaction between the electron donor and electron mediator using Fe₂O₃/Al electrode and Fe₃O₄/Al electrode as the working electrode in ammonium thiosulfate medium containing known concentrations of gold and copper ions. Cyclic voltammetry showed that the current peak due to gold reduction was de-

tected around -1.0 V vs Ag/AgCl, while current peaks due to copper reduction were not detected. Cyclic voltammogram and electrochemical impedance spectrum obtained in the experiments using high-purity iron oxides (magnetite and hematite) specimens as working electrodes also confirmed the selective reduction of gold. The results of SEM-EDX analysis for the cementation products confirmed that only gold was deposited on the surface of iron oxide electrodes. These results indicate that selective gold deposition is mainly due to the properties of iron oxides.

In Chapter 4, the mechanism for the selective gold deposition from ammonium thiosulfate solutions containing gold and copper ions on the iron oxide surface was discussed based on the results of zeta-potential measurements and investigations for the semiconductive properties of the iron oxides. The results of zeta potential measurements showed that the iron oxide surface was positively charged in ammonium thiosulfate solutions, implying that negatively charged gold thiosulfate complexes are electrostatically attracted to the iron oxide surface, but this is not for positively charged copper ammonium complexes. Investigations of the semiconductive properties of iron oxides showed that they were n-type semiconductors with a band gap energy of 1.0 eV for magnetite and 2.4 eV for hematite. The flat-band potentials were around -0.9 V for magnetite and -1.0 V for hematite. By configuring the energy band diagrams of magnetite and hematite, it was found that the redox potential of gold thiosulfate complexes is near the bottom edge of the conduction band for the iron oxides, which may cause a smooth electron transfer from iron-oxide to gold thiosulfate complexes in solutions. The redox potential of copper ammonium complexes was positioned in the band gap between conduction and valence bands, where the electron density is zero, and this may be caused by the limited electron transfer from the iron oxides to copper ammonium complexes in solutions.

Lastly, Chapter 5 summarizes the essential findings of this dissertation and its implications.