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Title	Development of a new method for gold recovery from waste mobile phones by combining physical separation and hydrometallurgical techniques using ammonium thiosulfate medium [an abstract of dissertation and a summary of dissertation review]
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Degree Grantor	北海道大学
Degree Name	博士(工学)
Dissertation Number	甲第13658号
Issue Date	2019-03-25
Doc URL	https://hdl.handle.net/2115/74070
Rights(URL)	https://creativecommons.org/licenses/by-nc-sa/4.0/
Type	doctoral thesis
File Information	Jeon_Sanghee_abstract.pdf, 論文内容の要旨



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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　Jeon Sanghee

学 位 論 文 題 名

Development of a new method for gold recovery from waste mobile phones by combining physical separation and hydrometallurgical techniques using ammonium thiosulfate medium
(物理選別とチオ硫酸アンモニウム溶液を用いた湿式製錬を組み合わせた廃携帯電話からの新しい金回収法の開発)

The life-cycle of mobile phones is probably the shortest among electronic devices, which leads to increase in the generation of waste mobile phones. Mobile phones are complex devices typically composed of a plastic frame, liquid crystal displays, and printed circuit boards (PCBs). PCBs contain a wide variety of materials including not only hazardous elements but also valuable metals like gold (Au). The average Au content in PCBs is several hundreds of times higher than natural ores, so the recycling of Au are needed for resources sustainability and conservation. The recovery of Au from electronic wastes typically requires two stages: (1) hand-dismantling to specifically target valuable components (e.g., PCBs), and (2) metallurgical treatments to extract and refine target metals. For waste mobile phones, hand-dismantling to recover PCBs is impractical due to their small sizes, so only batteries are removed, and the remainder is treated by metallurgical techniques. Pyrometallurgical treatment requires high initial capital and operational costs, so hydrometallurgical technique is a good alternative in areas without any pyrometallurgical facilities. In hydrometallurgy, Au is first leached in suitable aqueous solutions and then recovered by adsorption, precipitation or cementation. Cyanide and halides are widely used, but they are either toxic or highly corrosive. Among the available alternatives, ammonium thiosulfate is attractive because it is non-toxic, less corrosive, and highly selective for Au. Despite these many advantages, its use in real plant is still limited because of two drawbacks: (1) Au extraction is low at high solid-to-liquid ratio (S/L), and (2) acceptable methods to recover extracted Au ions are not yet established. This study investigated the leaching of Au in mobile phones and identified the reasons why Au extraction was low at high S/L. Moreover, a pretreatment flowchart was developed to separate interfering metal(s) from the Au-containing fractions and improved the gold leaching efficiency in ammonium thiosulfate medium. Finally, a novel method to recover extracted Au ions in ammonium thiosulfate medium was developed.

Chapter 1 gives the background, importance and objectives of the study.

In Chapter 2, previous studies on electronic wastes recycling were reviewed.

In Chapter 3, optimum leaching conditions for Au extraction were established. The parameters evaluated include S/L (0.1-1g/10ml), concentration of thiosulfate (0-3 M), cupric sulfate (0-10 mM), ammonium sulfate (2.5-250 mM), and ammonium hydroxide (5-500 mM), leaching time (0-75 h) at 25 ^{o}C (pH 9-10). The results showed that Au extraction increased with increasing shaking time, total ammonium, and cupric sulfate concentration. Increasing the concentration of thiosulfate also enhanced the extraction of Au but only up to 1M. Higher than this value, however, the extraction of Au decreased.

The optimum Au extraction was achieved under the following condition: 1 M of thiosulfate, 10 mM of cupric sulfate, 1 M of total ammonium concentration, 0.1 g/10 ml of S/L and 24 h of leaching time.

Chapter 4 identified the interfering metal(s) that caused cementation/precipitation of extracted Au ions during ammonium thiosulfate leaching. SEM-EDX observations of the leaching residues showed significant amounts of Au together with Cu and Al, implying that extracted Au ions were likely re-deposited onto these metals via reductive deposition. A more detailed elucidation of this phenomenon showed that although Cu or Al alone could recover Au ions via cementation, this process became more extensive when they were suspended together even though the amounts of the individual metals were very low. Chronoamperometry results in ammonium thiosulfate solutions using an Al working electrode also showed that Au ion cementation was dramatically enhanced when Cu powder was present in solution, and the bulk of Au was cemented on Cu powder rather than on the Al electrode. These results suggest that the coexistence of Cu and Al interfered with the extraction of Au at high S/L because of the enhanced re-deposition of extracted Au via galvanic interaction. For this reason, separation is needed to separate interfering metals like Cu and/or Al prior to leaching.

Chapter 5 identified the effects of physical separation of interfering metals on Au extraction, and practical physical treatment flowchart was proposed. Several studies that applied both physical and hydrometallurgical treatment have been conducted, but those studies did not evaluate how physical separation affects the leaching efficiency of Au. In this chapter, mineral processing techniques were applied, and the effects of these techniques on Au extraction were evaluated. The proposed flowchart includes the following: (1) pre-concentration (1st jig separation) of PCBs from coarse crushed products of mobile phones (+2–10 mm), (2) removal of Cu-/Al-containing parts by 2nd jig separation from the crushed products of PCBs recovered in the step (1), (3) ammonium thiosulfate leaching of Au. The fine fractions generated from coarse and fine crushing were treated by flotation and hybrid jig separation to separate Au containing parts. Au extraction was improved by about 11-fold.

In Chapter 6, the novel method to recover Au ions from ammonium thiosulfate leached solution using the synergistic effects of Al and activated carbon on Au cementation was developed. When only activated carbon or Al was present in a thiosulfate solution, Au recovery was negligible under the following conditions: 0.15 g of activated carbon or Al with 10 ml thiosulfate solution containing 100 mg/l of Au ions at 25 ^{o}C for 24 h with 120 rpm. When both 0.15 g of activated carbon and 0.15 g of Al coexisted in the solution, however, Au recovery significantly increased (over 99%) via two possible mechanisms. Firstly, nano-scratches on the oxide film of Al were formed by particle-particle collision during shaking. Fine particles of activated carbon attached to the nano-scratches most likely enhanced the transfer of electrons from Al to Au ions, which resulted in Au cementation on activated carbon. Secondly, the thin Al oxide film most probably acted as a dielectric material that accumulated electrons within the oxide film and donating them to the attached activated carbon on Al where Au ions could be deposited.

Finally, Chapter 7 gives the general conclusions of this study.