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Gold recovery from shredder light fraction of E-waste recycling plant by flotation-ammonium thiosulfate leaching



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ABSTRACT

This paper describes the recovery of gold (Au) from shredder light fraction (SLF) of a recycling plant by flotation and leaching. SLF is typically sent to landfills as waste, but it still contains substantial amounts of Au, and other metals like Cu and Fe. The SLF sample used in this study contains 0.003% of Au, 12% of Cu, and 10% of Fe. Flotation results showed that over 99% of Au and 50% of combustibles were recovered in froth while most of the base metals were recovered in tailing. SEM-EDX of froth products indicates that Au floated via two mechanisms: (1) flotation of Au-plated plastic particles, and (2) agglomeration of fine Au particles together with plastic particles due to kerosene-induced hydrophobic-hydrophobic interactions followed by the flotation of these agglomerated particles. Combustibles in froth/tailing were analyzed by ATR-FTIR, and the results showed that plastics in the froth were mostly sulfonated polystyrene (PS) and acrylonitrile butadiene styrene (ABS) while those in tailing were polyurethane (PU) and polyethylene terephthalate (PET). Contact angle measurements of plastic particles suggest that PS and ABS are more hydrophobic than PU and PET. Most of the base metals in the tailing had either bent or twisted shapes because they were mostly made up of wires. In flotation, these large and heavy particles are unaffected by bubbles and simply sink. Leaching results using ammonium thiosulfate solutions showed that Au extraction increased from 33 to 51% after flotation.

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1. Introduction

In 2016, electronic wastes (E-wastes) generation was around 44.7 million metric tons (Mt), equivalent to 6.1 kg per person, and is estimated to grow by about 4% until 2021 (Akcill et al., 2015; Lee et al., 2007). E-wastes generally contain various kinds of substances with diverse chemical compositions including not only valuable metals like gold (Au), copper (Cu), iron (Fe), and aluminum (Al) but also some hazardous elements and compounds (e.g., mercury and brominated flame retardants) (Ha et al., 2014; Mmereki et al., 2016; Surface Treatment Experts, 2017; You and Park, 2014). Among the various materials found in E-wastes, Au is the most valuable, usually accounting for over 70% of the total metal price (Park et al., 2017; You and Park, 2014). On average, 1 ton of E-waste containing about 0.025 wt% of Au is more than 25–250 times higher than the average Au ore (1–10 g/ton) (You

and Park, 2014). This means that Au recovery from E-wastes is profitable and is in fact currently being exploited by government and private companies.

Recycling of end-of-life electronic machines typically involves several stages: (1) manual dismantling to specifically target large parts that are either valuable or hazardous, (2) shredding to reduce volume, (3) liberation of valuable components from the nonvaluable materials, (4) concentrating to increase the amount of valuable materials in the end-products, (5) extraction to recover metals from the end-products utilizing either pyrometallurgical or hydrometallurgical technique, and (6) refining (Lee et al., 2007; Sarvar et al., 2015). Almost all recycling plants employ some kind of shredding stage to crush E-wastes, which is typically a dry-type operation. The shredded fraction goes to air separation where fine particles are collected together with relatively large (up to 10 cm) but light materials as shredder light fraction (SLF) (Ad Rem, 2013; Ogunniyi and Vermaak, 2009). SLF is commonly classified as wastes and is disposed of in special landfill sites like other hazardous industrial wastes or transported to power generation plants (Ad Rem, 2013; Park et al., 2018; Tabelin and Igarashi, 2009; Tabelin et al., 2012; Tabelin et al., 2014). However,





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disposal of SLF has several drawbacks: (1) loss of valuable materials, and (2) release of hazardous elements like mercury and heavy metals (Yang et al., 2011). The relative amounts of valuable metals vary in SLF depending on the type of E-wastes processed by recycling plants, but they are typically high enough for metals recovery to be economically viable. For example, SLF generated in a recycling plant in Japan contains up to about 0.004 wt% (40 ppm) of Au, 11 wt% of Cu, and 3 wt% of Fe (Ito et al., 2016). This means that SLF should be considered as a resource rather than as wastes.

Several authors have also pointed out that E-wastes and automotive shredder residues (ASR) are important resources to recover metals and combustible materials (Francesco et al., 2015; Yang et al., 2011). Previous works on SLF and ASR, for example, have succeeded in the recovery of oil from combustible materials by pyrolysis (Andersson et al., 2012; Galvano et al., 2001; Roy and Chaala, 2001; Santini et al., 2012). Unfortunately, an efficient way to recover both valuable metals, such as Au, and combustible materials is still not available. To the best of our knowledge, this is the first paper that tackles both separation of combustibles and metallic fractions as well as the recovery of Au from SLF.

To recover Au and separate combustibles and base metals in SLF obtained from an E-waste recycling plant, flotation, a commom physical separation technique, was applied. In mineral processing, flotation is typically applied to concentrate target minerals (e.g., Cu, lead, zinc and tin minerals) based on differences in their physico-chemical properties brought about by the addition of various types of flotation reagents (Feng et al., 2017a, 2017b, 2018; Feng and Wen, 2017; Wills and Napier-Munn, 2006). SLF predominantly contains various combustible materials (mainly plastics) and metals, so they could be separated because the former are typically hydrophobic (i.e., water-attracting) while the latter are hydrophilic (i.e., water-repelling) (Das et al., 2009). In other words, flotation could potentially recover most of the combustible fraction in the froth products while metallic fractions end up in the tailing.

In the present study, separation of combustibles and metallic fractions as well as the recovery of Au from SLF were carried out by flotation. Kerosene was used as a collector to enhance the hydrophobicity of plastic particles and the effects of flotation time were investigated. Recovery of combustible materials was determined by ashing-gravimetric method while those of Au and coexisting metals were quantified by inductively coupled plasma atomic emission spectroscopy (ICP-AES) after ashing and aqua regia digestion of the ash. The froth and tailing products were also examined by high-resolution digital microscopy, scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX) and attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy to understand how Au, metallic particles, and plastics behaved during flotation. Finally, leaching was conducted to compare Au extraction with and without flotation and determine whether the compounds used during flotation have detrimental effects on Au extraction.

2. Materials and methods

2.1. Characterization of shredder light fraction sample

The SLF sample was obtained from a recycling plant in Japan that handles end-of-life industrial wastes (e.g., photocopierprinters, and automated teller machine (ATM)) as well as home appliances including computer (Supplementary Fig. 1). Visual inspection of the SLF sample showed that it is a mixture of wood, various plastics, papers, fine particles of metals, and various electronic components. Preliminary experiments showed that most of the large combustible materials (e.g., plastics) could be separated from the fine fraction containing the bulk of metallic compo-

nents including 0.003% of Au by screening (2 mm aperture screen) (Supplementary Fig. 2), so this study focused on the recovery of Au as well as the separation of combustibles and base metals from the less than 2 mm size fraction. Size distribution of the feed was measured by sieving while liberation analysis of each size fraction was carried out using a high-resolution digital microscope (VHX-1000, Keyence Corporation, Japan). In practice, liberation is considered sufficient when >90% of target minerals and gangue minerals exist as free particles. The chemical composition of the feed material (<2 mm SLF) was determined by a special ashing technique and aqua regia digestion (Inorganic Ventures, 1985; Yamane et al., 2011). The ashing method was developed by Tomita et al. (1983) to prevent the volatilization of Au during thermal treatment of PCBs, and involves slow heating of the sample to 500 °C for 3 h using an ashing furnace (Ishizuka Denki Seisakusho, Japan) followed by the continued burning of the sample at this temperature for 5 h (Tomita et al., 1983). After aqua regia digestion of the resulting ash, the leachate was filtered through 0.2 µm membrane filters (Sartorius AG, Germany), and the filtrates were analyzed by ICP-AES (ICPE-9820, Shimadzu Corporation, Japan) (margin of error = ±2%).

2.2. Flotation

The flotation experiments were conducted with 20 g of the sample and 400 ml of distilled water in a batch-type mechanical flotation cell (Agitator-type, FT-1000, Heiko, Japan) under the following conditions: impeller speed of 1000 rpm, air flow rate of 0.75 l/min, and 40.5 µl/l of methyl isobutyl carbinol (MIBC) as frother as well as 405 g/ton of kerosene (405 g of kerosene/ton of SLF) as collector. Plastics are inherently hydrophobic similar to coal, so kerosene was selected as a collector since it is typically utilized in coal cleaning. Moreover, it is relatively cheap compared with other compounds used as collectors (Yang et al., 2011). In the flotation experiments, the effects of time (0-10, 10-30, 30-60, 60-120, 120-180, 180-200 s) and kerosene dosage were investigated. Materials in the froth and tailing products were identified by microscopy and analyzed by SEM-EDX (SSX-550, Shimadzu Corporation, Japan) while their combustible and chemical compositions were quantified by the procedure outlined in the previous subsection.

2.3. Characterization of plastics in froth and tailing

Surface and chemical properties of plastic particles were identified by contact angle measurements coupled with ATR-FTIR spectroscopy (FT/IR-6200 HFV and ATR Pro One attachment equipped with a diamond prism, Jasco Analytical Instruments, Japan). Captive bubble method with image analysis was employed to measure contact angles (Supplementary Fig. 3), which was done by fixing the samples in a holder and then immersed into distilled water prior to the introduction of a bubble on the surface of the sample using a microliter syringe (DS 500/GT, Hamilton, Japan). The contact angle was measured using a high-resolution microscope with image analysis capability.

2.4. Leaching

Ammonium thiosulfate solution was utilized in the leaching experiments because it is non-toxic and less corrosive compared with cyanide and halide solutions. The leaching experiments were conducted by mixing 1 g of feed or froth products and 10 ml of solution containing 1 M Na₂(S₂O₃), 0.5 M NH₄OH, 0.25 M (NH₄)₂SO₄ and 10 mM CuSO₄ in 50 ml Erlenmeyer flasks (these conditions were selected based on our preliminary experiments (Supplementary Fig. 4)). The suspensions were shaken in a

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