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# Recovery of precious metals from low-grade automobile shredder residue: A novel approach for the recovery of nanozero-valent copper particles

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## ABSTRACT

The presence of precious metals (PMs) in low-grade automobile shredder residue (ASR) makes it attractive for recycling. This study investigated the leaching and recovery characteristics of two PMs (Cu and Ag) and two heavy metals (Mn and Co) from ASR. The effects of H<sub>2</sub>O<sub>2</sub>, leaching temperature, liquid to solid (L/S) ratio, and particle size on metal leaching were determined in an aqueous solution of 0.5 M nitric acid. The metal leaching rate was increased with increasing nitric acid concentration, amount of H<sub>2</sub>O<sub>2</sub>, L/S ratio and temperature. The leaching kinetics was analyzed by using a second-order reaction model. In the analysis of leaching kinetics, the metal leaching data were well fitted ( $R^2 \geq 0.99$ ) with the second-order reaction model. The activation energy (kJ/mol) for metal leaching was 39.6 for Cu, 17.1 for Ag, 17.3 for Mn and 29.2 for Co. Metal recovery was carried out by fractional precipitation with the addition of advanced Fenton's reagent. Metal recovery efficiency was increased to 99.95% for Cu, 99.8% for Mn, 90.0% for Ag and 96.46% for Co with the advanced Fenton's reagent. In particular, a novel finding of the PM recovery is that Cu can also be recovered directly from the leachate of ASR in the form of zero-valent copper (ZVC) nanoparticles (NPs). Hydrometallurgical recovery of the metals from ASR using nitric acid is highly efficient.

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## 1. Introduction

Precious metals (PMs) are used increasingly in the fabrication of electronic devices due to their special electric conductivity. The recovery of PMs from the electronic waste by different techniques have already been done by many researcher (Zhang et al., 2012; Ruan et al., 2014; Ghosh et al., 2015; Hadi et al., 2015). However, recovery of PMs from low-grade automobile shredder residue (ASR) is rather limited. The ASR fraction >5.0 mm can be used for energy generation, however ASR size <5 mm is neither reused nor recycled, and finally dumping into landfills, called as low grade ASR (Singh and Lee, 2015). The coarse fraction has the high volatiles, lowest ash content and the highest calorific value (15–30 kJ/kg), resulting highly recommend for energy generation. On the other hand, the fine fraction (low grade ASR) has low volatile, higher ash content and a lower calorific value (11–21 MJ/kg). The fine fraction is not good for incineration process, so it can be

recommended for energy generation (Morselli et al., 2010; Vermeulen et al., 2011; Cossu et al., 2014; Singh and Lee, 2015).

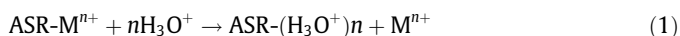
ASR containing PMs is dumping into landfills. Recovering PMs from ASR is beneficial for conserving PM resources and protecting the environment. Therefore, it is very important to discover new and effective methods for recovering PMs from ASR. Hydrometallurgical recovery of PMs from low-grade ASR is a possible alternative because of its suitability for small-scale applications and reduction of environmental impacts due to reduced emissions of dangerous gases and dust (Cui and Zhang, 2008; Tuncuk et al., 2012; Kubonova et al., 2013). ASR has been considered hazardous waste due to its high heterogeneity and variable composition of various toxic metals, organic pollutants, glass, and plastics (Granata et al., 2011). ASR contributes about 20–25% of the total volume of end-of-life vehicles (Kurose et al., 2006; Jung et al., 2007). Despite containing PMs, most low-grade ASR is simply and directly disposed of into landfills (Oshitani et al., 2003; Donaja et al., 2010). Disposal of hazardous residues (ASR, E-waste) can cause serious health issues and have a detrimental influence on the environment, such as water contamination due

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to the leaching of processed and unprocessed wastes (Cayumil et al., 2014).

PMs are present in elemental forms and/or alloys in ASR and thus oxidizing conditions are often required for leaching of PMs. Nitric acid is a distinctive and strong oxidizing agent as it combines with hydrogen peroxide. Nitric acid is therefore a competent leaching agent for the treatment of ASR (Bas et al., 2014). Nitric acid is also extensively used in combination with hydrochloric acid in the form of aqua-regia for the determination of metals in ASR (Granata et al., 2011). Nitric acid alone is a highly capable, selectively extracting agent for the leaching of metals such as Cu, Ni, Pb and Ag (Mecucci and Scott, 2002; Kinoshita et al., 2003; Le et al., 2011; Bas et al., 2014). Nitric acid is a strong Bronsted acid, and thus is completely dissociated in aqueous solution and becomes a good source of hydronium ions. The metal leaching from ASR by inorganic acids is based on the exchange of metal ions with hydronium ions followed by their solubilization, as described in Eqs. (1) and (2). Determination of the leaching efficiency of PMs from ASR is very important because it can affect the feasibility of selected leaching methods (Deng et al., 2009).



The application of Cu<sub>2</sub>O and nano zero-valentcopper (nZVC) has become the most promising innovative technology for treating waste water organic contaminants (Dong et al., 2014; Huang et al., 2012, 2013). Nano ZVC has high conductivity and malleability and it allow for the reduction of O<sub>2</sub> into H<sub>2</sub>O<sub>2</sub>. Therefore, nZVC might be a sustainable O<sub>2</sub> activator for the degradation of organic contaminants (Dong et al., 2014). Nano ZVC has also the capability to initiate many organic reactions and induce DNA degradation (Jose et al., 2011; Ren et al., 2013). It has been used as a catalyst for the degradation of dichloromethane (Huang et al., 2012, 2013) and azo-contaminates (Dong et al., 2014). ZVC and Cu<sub>2</sub>O have also been used as catalysts for the decoloration of methyl orange (Zhang et al., 2011). Therefore, the recovery of Cu in the form of Cu<sub>2</sub>O and ZVC nanoparticles (NPs) from ASR useful for treating organic contaminants (methyl orange, dichloromethane, azo dyes, etc.) in waste water and reducing the toxicity of ASR for its safe disposing into landfills.

Nano ZVC was directly synthesized from an aqueous solution of metals extracted from ASR. The twin objectives of the study were to recover effectively two PMs (Cu and Ag) and two metals (Mn and Co) from low-grade ASR, and to recover Cu in the form of Cu<sub>2</sub>O and ZVC NPs from the leached liquor of ASR. In this study, nitric acid was used to recover PMs from ASR.

## 2. Materials and methods

In order to obtained two PMs (Cu and Ag) and two metals (Mn and Co) from ASR, this study developed a procedural sequence for leaching and recovery, as shown in Fig. 1.

### 2.1. Preparation of ASR materials

An ASR sample was collected from the shredder plants of Steel SCRAP Ulsan factory, Nam-gu, Ulsan, Korea. The ASR sample (approximately 100 kg) was collected from the different places of heap (near shredder residue plant) as shown in Fig. 2. After proper mixing, sample was sub-divided at least three times using the long pile principle. Approximately 20 kg sample was brought to laboratory for analyses. The sample was sieved through a 4.75 mm sieve, and the bigger particles (>4.75 mm) were not used for the study. The remaining ASR particles were sieved to obtain five different

size fractions: A: 4.75–2.0 mm; B: 2.0–1.19 mm; C: 1.19–0.425 mm; D: 0.425–0.250 mm and E: 0.250–0.0 mm. Different fractions of the sieved ASR were dried in a hot air oven (C-DF forced convectional drying oven, Chang Shin Scientific Co.) at 105 °C for 24 h before the leaching study was conducted.

### 2.2. Leaching experiment

Batch study was performed for metal leaching in a series of 250 mL covered conical flasks using a temperature-controlled water bath shaker (HST-205SW) at 150 rpm. The metal leaching from ASR was performed at different HNO<sub>3</sub> concentrations (0.5 M, 1.0 M, 2.0 M and 3.0 M), liquid to solid (L/S) ratios (10, 25, 50 and 100 mL/g), temperatures (303, 313, 323 and 333 K) and particle sizes (4.75–2.0, 2.0–1.19, 1.19–0.425, 0.425–0.250 and 0.25–0.0 mm). The influences of different H<sub>2</sub>O<sub>2</sub> concentrations on PM leaching were investigated with 0.5 M HNO<sub>3</sub>. The size fractionized ASR samples were leached for various times (5, 15, 30, 60, 90, 120 min) to examine the effect on leaching efficiency. The samples were centrifuged, filtered through filter paper (0.45 μm) and the metal concentrations in the filtrate solutions were determined. Each experiment was repeated three times and the results were averaged. The metal leaching efficiency was calculated by using the following formula (Eq. (3)) (Hong et al., 2000):

$$\text{Leaching efficiency (\%)} = (C_L/C_T) \times 100 \quad (3)$$

where  $C_L$  is concentration of metals in liquid (mg/kg) and  $C_T$  is total concentration of metals obtained after strong acid digestion (mg/kg).

### 2.3. Metal recovery from the leachate

For the metal recovery from the leachate, a fractional precipitation method was used. The principle of the fractional precipitation is based on the inconsistent solubility of selected metals (Chen et al., 2014). As shown in the sequence in Fig. 1, a leaching and recovery of metals was carried out with different chemical reagents. The precipitation experiments were conducted with a Jar test method (Sthiannopkao and Sreesai, 2009; Fu et al., 2012). Fractional precipitation experiments were performed in a 500 mL beaker with 200 mL of actual liquid volume. The pH was adjusted by using 0.1 M NaOH and 1.0 M HCl with stirring at 80 rpm for 10 min using a temperature-controlled magnetic stirrer (GLHF-G, Global lab). Then the solid precipitates were allowed to settle for 20 min. The precipitation chemical study was carried out at 20 ± 2 °C. The recovery of selected metals was enhanced by advanced Fenton system (Fu et al., 2012). During each successive step, 0.125 g/L zero-valent iron (ZVI) concentration and 0.5% (V/V) H<sub>2</sub>O<sub>2</sub> (conc. 30%) were added to the acidic solution (pH 1.5–3.0) containing metals extracted from ASR (Fu et al., 2012). The mixture was stirred for 20 min for the reaction with H<sub>2</sub>O<sub>2</sub> and nZVI. For the precipitation of metals, the required pH was adjusted for each metal and then the Jar test method was conducted. Each precipitation test was replicated three times. After each precipitation step, the solution was centrifuged at 4000 rpm for 10 min and then filtered with a 0.45 μm filter. The resulting filtrate was used for the next precipitation process until the last targeted metal was recovered. From each filtrate, 10 mL solution was collected for analyzing the residual metal concentration. Metal recovery was calculated by using the following Eq. (4) (Fu et al., 2012):

$$\text{Metal recovery (\%)} = [(A - B)/A] \times 100 \quad (4)$$

where  $A$  is the concentration of a metal after leaching and  $B$  is the residual concentration after precipitation.

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