



Reduction of environmental availability and ecological risk of heavy metals in automobile shredder residues



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ABSTRACT

The present study investigated the recovery, reduction of environmental availability and ecological risk of heavy metals extracted from automobile shredder residue (ASR) using water with pH 2. The recovery rate of heavy metals from ASR increased with increasing temperature and L/S ratio at pH 2. The heavy metal recovery was found to be greater in the smallest size fraction than in the larger fractions. The residues remaining after extraction were analyzed for functional groups, morphology, toxicity characteristic leaching procedures (TCLP) and mobility factors (MFs) of the heavy metals. The TCLP test results revealed Cd concentrations below the threshold limits in all ASR fractions, whereas Pb concentrations exceeded the threshold limit in all ASR fractions, with the exception of the largest fraction. The MFs of all selected heavy metals were reduced after extraction. In the analysis of the risk assessment code (RAC), Zn, Cu and Cd were reduced from the high risk category into either low risk or medium risk categories after recovery of the heavy metals, whereas Pb remained under the high risk category. This confirmed that distilled water at pH 2 is not efficient for reducing Pb toxicity to the extent necessary for safe landfilling of ASR. Therefore, a stronger acid is required for the higher recovery of Pb and reduction of its toxicity before ASR is landfilled.

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1. Introduction

Leachate is generated through landfilling of heavy metals-contaminated wastes such as municipal solid waste and automobile shredder residue (ASR). It is considered a major source of metal contamination in nearby aquatic environments (Salem et al., 2014). Heavy metal release under acidic conditions is one of the major environmental concerns for the landfilling of automobile shredder residue (ASR) under acidic conditions. The heavy metals in ASR originate from residual metal pieces, solders, plasticizers and paints (Kurose et al., 2006). The heavy metals may leach out from ASR to pollute soil, ground water and surface water; these metals can transfer to the ecosystem and the food chain, posing a hazard to human health (Shaheen and Rinklebe, 2015). Thus, the leaching of heavy metals from ASR has raised public concern, with ASR classified as a hazardous waste that must be disposed of in controlled landfill sites after proper treatment (Gonzalez-Fernandez et al., 2008). Heavy metals in water-soluble fractions can leach out easily and become bioavailable in the environment (Liu et al., 2008; Singh and Kalamdhad, 2013a,b; Singh and Kalamdhad, 2014a). More attention

should be given to the weakly bonded exchangeable (EXCH) and carbonate bound (CARB) fractions as they can equilibrate with the aqueous phase following changes in the ionic concentration and pH of the soil and consequently, become rapidly bioavailable (Sundaray et al., 2011; Singh and Kalamdhad, 2013a,b; Shi et al., 2013). Determination of the specific quantity of heavy metals in ASR can be performed using a risk assessment code (RAC) based on the proportion of these two fractions (EXCH and CARB) compared to the total concentration of each metal obtained after strong acid digestion of ASR (Huang et al., 2011). The toxicity or leachable fractions of heavy metals in ASR can be reduced by extraction with water, which is very important for reducing environmental risk.

Therefore, the safe disposal of ASR into landfills necessitates the extraction/recovery of heavy metals from ASR using an economically suitable extraction agent. Extraction of heavy metals from ASR using water is regarded as environmentally friendly and has been employed for the removal of heavy metals from fly ash generated during municipal solid waste incineration. The toxicity and leaching characteristics of heavy metals can be greatly reduced after extraction with water (Wang et al., 2001). Under acidic conditions such as pH 2, heavy metals in landfills are easily leached out from ASR during the rainy season. Therefore, the objectives of the present study were to recover heavy metals (Zn, Cu, Pb and Cd) from ASR by extraction using water at pH 2 and to reduce the

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environmental toxicity risk of heavy metals in ASR before disposal into landfill sites.

2. Materials and methods

2.1. Characterization of ASR

A 20 kg ASR sample was collected from the Steel SCRAP Ulsan factory shredder plant at Nam-gu, Ulsan, Korea. The sample was passed through a sieve of 4.75 mm, and the larger particles (>4.75 mm) were discarded. The remaining particles were sieved again to obtain five different grain-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. An atomic absorption spectrometer (AAS) (Varian Spectra 55) was used to analyze the Zn, Cu, Pb and Cd concentrations in the different fractions of ASR.

2.2. Experimental setup

Batch study was performed in a series of 250 mL covered conical flasks using a temperature-controlled water bath shaker (HST-205SW) set at 150 rpm. The extraction of heavy metals from ASR was performed at different pHs (2, 4, 6, 8, 10 and 12), L/S ratios (10, 25, 50 and 100 mL g⁻¹), temperatures (25, 35, 45 and 55 °C) 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 experimental extraction time was 120 min and the samples were withdrawn at various time intervals for analysis of the heavy metals extracted. The samples were centrifuged and filtered through filter paper (0.45 μm) and the filtrates were used for heavy metal analysis. The pH of the deionized water was maintained by adding 1.0M HCl and NaOH. The final pH of the extracted solution was measured. Each experiment was repeated three times and the results were averaged. Heavy metal recovery

was calculated using the following formula (Hong et al., 2000): Heavy metal recovery = [(Concentration of metals in extraction liquid)/(total contents of metals obtained after acid digestion) × 100].

Instrumental analysis

The solid residues obtained under specific conditions (temperature 25 °C, stirring speed 150 rpm, solution pH 2, L/S 50 mL g⁻¹ and extraction time 60 min) were chosen for scanning electron microscopy (SEM) analysis (Hitachi S-4700), Fourier transform infrared (FTIR) spectroscopy (Nicolet Nexus 470 FTIR spectrometer), and X-ray fluorescence (XRF) spectrometry (S8 Tiger, Bruker, Germany) and X-ray diffraction (XRD) analysis. SEM analysis was performed to characterize the morphology of the ASR before and after heavy metals recovery. FTIR spectroscopy of the ASR samples was performed in the region 4500–500 cm⁻¹ to analyze any changes in functional groups after heavy metals recovery. The XRD patterns were recorded on a Bruker AXN in the 2θ range of 10–80° using Cu-Kα radiation (λ = 1.5418 Å) to check the crystallinity of the ASR before and after the recovery of metals. A XRF spectrometry was performed to determine the elemental composition of the ASR samples before and after heavy metals recovery.

2.4. Toxicity characteristic leaching procedure (TCLP) test and sequential extraction

After the recovery process, the residue material was oven dried at 105 °C for 24 h, and then ground and desiccated until testing began. The residues obtained after heavy metal recovery were subjected to a TCLP test (USEPA 1311, 1992) and to the sequential extraction method developed by Tessier et al. (1979). For the TCLP

Table 1

Total metal content and maximum recovery of heavy metals over a range of pH, temperature, L/S ratio and grain fractions.

| Variation of parameters | Heavy metals | | | | | |
|--|--------------|----------------------|-------------|----------|--------|------|
| | Zn | Cu | Pb | Cd | | |
| Total heavy metals (mg kg⁻¹) | | | | | | |
| 1.0 g of the ASR sample | A | 634 ± 152 | 3.189 ± 100 | 375 ± 25 | 42 ± 1 | |
| digested with 20 mL of H ₂ SO ₄ and | B | 1.164 ± 44 | 3.723 ± 64 | 345 ± 25 | 37 ± 2 | |
| HClO ₄ (5:1) mixture in a closed Teflon vessel | C | 1.546 ± 305 | 2.504 ± 117 | 370 ± 30 | 38 ± 4 | |
| | D | 1.657 ± 6 | 2.673 ± 14 | 350 ± 40 | 37 ± 2 | |
| | E | 4.823 ± 71 | 3555 ± 57 | 770 ± 90 | 63 ± 9 | |
| | | Maximum recovery (%) | | | | |
| pH | | 2 | 14.89 | 3.44 | 1.75 | 2.06 |
| | | 4 | 1.73 | 0.35 | 0.18 | 0.24 |
| (L/S-50 mL g ⁻¹ , temperature – 25 °C, agitation speed – 150 rpm and grain size fraction <0.25 mm) | | 6 | 1.76 | 0.34 | 0 | 0 |
| | | 8 | 1.42 | 0.36 | 0 | 0 |
| | | 10 | 1.95 | 0.44 | 0 | 0 |
| | | 12 | 1.22 | 0.52 | 0 | 0 |
| Temperature (°C) (L/S-50 mL g ⁻¹ , pH 2, agitation speed – 150 rpm and grain size fraction <0.25 mm) | | 25 | 14.89 | 3.44 | 1.75 | 2.06 |
| | | 35 | 4.34 | 3.74 | 2.27 | 1.93 |
| | | 45 | 4.66 | 3.89 | 2.86 | 1.88 |
| | | 55 | 4.26 | 4.34 | 3.29 | 1.9 |
| L/S ratio (mL g⁻¹) (pH 2, temperature – 25 °C, agitation speed-150 rpm and grain size fraction <0.25 mm) | | 10 | 11.38 | 0.19 | 0.23 | 0.71 |
| | | 25 | 13.65 | 0.79 | 0.46 | 1.47 |
| | | 50 | 14.89 | 3.44 | 1.75 | 2.06 |
| | | 100 | 29.37 | 20.89 | 41.56 | 4.21 |
| Grain size fractions (pH 2, L/S-50 mL g ⁻¹ , temperature – 25 °C, agitation speed – 150 rpm) | A | 3.538 | 3.04 | 1.02 | 0.78 | |
| | B | 3.88 | 5.23 | 1.22 | 0.88 | |
| | C | 3.9 | 5.41 | 1.28 | 0.87 | |
| | D | 5.3 | 9.21 | 1.58 | 0.89 | |
| | E | 14.89 | 3.44 | 1.75 | 1.31 | |

Note: A: ASR size range 4.75–2.0 mm; B: size range 2.0–1.19 mm; C: size range 1.19–0.425 mm; D: size range 0.425–0.25 mm; E: size <0.25 mm.

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