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Reduction of heavy metals in residues from the dismantling of waste electrical and electronic equipment before incineration

Yu-Yang Long^a, Yi-Jian Feng^a, Si-Shi Cai^a, Li-Fang Hu^b, Dong-Sheng Shen^{a,∗}

a Zhejiang Provincial Key Laboratory of Solid Waste Treatment and Recycling, School of Environmental Science and Engineering, Zhejiang Gongshang University, Hangzhou 310012, China

b College of Quality and Safety Engineering, China Jiliang University, Hangzhou 310018, China

• The highest metal reduction occurs at a 2.36 mm sieving size.

• Washing promotes heavy metal recycling without secondary pollution.

• Sieving and washing are environmentally friendly pretreatments for WEEE wastes.

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ABSTRACT

Residues disposal from the dismantling of waste electrical and electronic equipment are challenging because of the large waste volumes, degradation-resistance, low density and high heavy metal content. Incineration is advantageous for treating these residues but high heavy metal contents may exist in incinerator input and output streams. We have developed and studied a specialized heavy metal reduction process, which includes sieving and washing for treating residues before incineration. The preferable screen aperture for sieving was found to be 2.36 mm (8 meshes) in this study; using this screen aperture resulted in the removal of approximately 47.2% Cu, 65.9% Zn, 26.5% Pb, 55.4% Ni and 58.8% Cd from the residues. Subsequent washing further reduces the heavy metal content in the residues larger than 2.36 mm, with preferable conditions being 400 rpm rotation speed, 5 min washing duration and liquidto-solid ratio of 25:1. The highest cumulative removal efficiencies of Cu, Zn, Pb, Ni and Cd after sieving and washing reached 81.1%, 61.4%, 75.8%, 97.2% and 72.7%, respectively. The combined sieving and washing process is environmentally friendly, can be used for the removal of heavy metals from the residues and has benefits in terms of heavy metal recycling.

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1. Introduction

The waste electrical and electronic equipment (WEEE) dismantling industry has grown in many developing Asian countries, because of the relatively poor supervision of imported waste in these countries and the increased demand for metal resources [\[1–3\].](#page--1-0) Compared with traditional mineral mining, recycling of metals from WEEE is simpler with greater energy-savings $[4]$. WEEE could be considered to be a type of urban mine with millions oftons of overseas WEEE being imported by developing countries every year. Taizhou, the largest WEEE dismantling base in East China, for example, imports approximately 2 million tons of WEEE annually

[http://dx.doi.org/10.1016/j.jhazmat.2014.02.048](dx.doi.org/10.1016/j.jhazmat.2014.02.048) 0304-3894/© 2014 Elsevier B.V. All rights reserved. from Japan, Korea, Europe and North America. This accounts for approximately 20% of the total WEEE import into China, with the amount of copper recycled being close to the production from Dexing Ore, the largest open pit copper ore mine in Asia [\[5\].](#page--1-0)

Although the WEEE dismantling industry yields high margins, it also poses significant environmental and health risks. A number of environmental pollution events have occurred in areas where WEEE dismantling is conducted, because of the high toxic materials content in WEEE and because unsophisticated treatment processes are adopted, such as burning in open fields and washing with acid [\[6–11\].](#page--1-0) Pollution from the WEEE dismantling industry can be controlled by eradicating household dismantling workshops and building enclosed WEEE dismantling bases. Despite these measures, current outputs of residues from the dismantling process are significant and problematic in WEEE dismantling areas. During the dismantling process, materials such as plastic cases,

[∗] Corresponding author. Tel.: +86 571 88832369; fax: +86 571 88832369. E-mail address: shends@zju.edu.cn (D.-S. Shen).

air-conditioning screens and jacketed electric cable are removed. Besides, a certain amount of industrial solid waste from overseas such as paper from packaging, scrap rubber, foam plastic and soil become mixed in with the WEEE during importation. These two kinds of wastes are unwanted because of their low recycling value and are therefore abandoned. These unwanted residues account for approximately 2% of the total imported amount of WEEE $[5]$, i.e., approximately 0.2 million generated in China. According to the WEEE import share of China (China receives approximately 90% of the total amount of WEEE imported to Asia [\[1\]\),](#page--1-0) about 0.22 million tons of dismantling residues may generated Asia.

These residues are non-biodegradable and are disposed of into municipal solid waste (MSW) landfills without volume reduction or other treatment. According to our previous work, landfill is not feasible for the disposal of dismantled residues because they have the potential to release large quantities of heavy metals [\[5\].](#page--1-0) WEEE dismantling residues can reduce leachate quality and there is a risk that they release heavy metals $[12]$.

Incineration, a promising disposal method for solid waste because of its volume reduction effect, sterilization and energy recycling [\[13–15\],](#page--1-0) may be a good choice for residues disposal. The high calorific value of WEEE residues makes their incineration feasible with the exception of the heavy metals mentioned above. The concentrations of Cu, Zn, Pb, Ni and Cd in these residues are 3549 mg kg⁻¹, 4575 mg kg⁻¹, 5331 mg kg⁻¹, 255 mg kg⁻¹ and 61 mg kg−1, respectively, according to the writer's previous investigation on 24 dismantling factories [\[5\].](#page--1-0) The contents of Cu, Zn, Pb, Ni and Cd are more than 5010 mg kg⁻¹, 9925 mg kg⁻¹, 10717 mg kg⁻¹, 383 mg kg⁻¹ and 32.4 mg kg⁻¹, respectively, in the bottom ash, and 2592 mg kg−1, 13851 mg kg−1, 10054 mg kg−1, 179 mg kg−¹ and 301 mg kg−1, respectively, in the fly ash when incinerated according to the writer's previous work [\[16\].](#page--1-0) These incineration products are metal-rich and difficult or expensive to solidify or stabilize before disposal [\[17–21\].](#page--1-0) Therefore, some environmentally friendly pretreatment strategies for heavy metal reduction should be implemented to decrease the heavy metal input to the incinerator. High efficiency pretreatment can decrease the risk of heavy metal release after incineration. Alternatively, the removed heavy metals could be collected as a metal-rich material for further metal recycling. An analysis of the source of heavy metals in the WEEE dismantling residues shows that these seem to concentrate in small particles and fragments during the WEEE dismantling cutting process [\[22–26\].](#page--1-0) Separation of the metal sources from the dismantled residues could reduce heavy metal contents significantly during incineration. In this research, a two-step pretreatment process strategy including sieving and washing was studied, to reduce and possibly recycle the heavy metal content from the residues before incineration.

2. Materials and methods

2.1. Materials

In order to obtain the representative sample of dismantling residues, approximately 360 kg of residue samples were collected from an incineration plant (120 t d^{-1} , burn dismantling residues from WEEE only) according to the Chinese industrial refuse sampling standard. Inert fractions including stone and ceramics were removed manually after sampling. Residues samples were then crushed to smaller than 5 cm, mixed homogeneously and quartered to produce subsamples. These subsamples were crushed to smaller than 8 mm and then to smaller than 0.5 mm using a shearing grinder (SM 2000, Retsch, Germany) to provide representative input residues samples. The 5-cm-sized residues (the mixture of

packaging paper, cable jacket, debris of WEEE, rubber and foam, soil, etc.) were used for the heavy metal removal test.

2.2. Heavy metal removal test

2.2.1. Sieving

The 5 cm residues (5 kg) were passed through a set of sieves (11, 4.75, 3.35, 2.36, 0.85, 0.50, 0.15 mm) using manual horizontal vibration. Vibration was continued until there was no obvious change in material in each sieve (approximately 3 min vibration required). The fraction in each sieve was weighed. Residues smaller than 11 mm and greater than 0.15 mm were ground to smaller than 0.15 mm by a jaw crusher (BB 51, Retsch). Residues larger than 11 mm were crushed to smaller than 0.5 mm using a shearing grinder (SM 2000, Retsch). Sieving experiments were carried out in triplicate.

2.2.2. Washing

Sieving can separate out caducous particles, however, a certain amount of small particles remain attached to the residues, possibly as a result of electrostatic attraction or adherence. A washing process was therefore used after sieving for further heavy metal removal.

Washing experiments were conducted with distilled water in a columnar barrel (20 L) with blade agitator and 8 mm sieve in the middle of the barrel (to prevent the settling out of large material).

Single-factor washing experiments were conducted at 100, 150, 200, 250, 300, 350 and 400 rpm for 1, 2, 3, 5, 7 and 10 min and liquid-to-solid ratios (L/S) of 20:1, 30:1, 40:1, 50:1, 60:1, and 70:1. Orthogonal experiments were then conducted based on results from the single-factor experiments. After washing, residues that had been intercepted by the 8 mm screen were collected, ovendried at 105 ◦C, weighed and ground (SM 2000, Retsch) to smaller than 0.5 mm for further analyses. Sediments were collected, ovendried at 105 ◦C and ground to smaller than 0.15 mm with a mortar for further analysis. Each washing test was carried out in triplicate.

2.3. Loss of ignition (600 \degree C)

In order to indirectly reflect combustibility of the dismantling residues, the LOI $_{600}$ \circ tests were conducted on the samples of different particle sizes [\[27\].](#page--1-0) Representative sample of each particles size (5.00 g) was put in crucibles, followed by ignition in muffle furnace under 600 °C for 3 h. After ignition, the residual weight of each sample was weighed, and the LOI_{600} \circ was calculated as follow:

$$
LOI_{600^{\circ}C} = \frac{(5.00 - RW) \times 100\%}{5.00}
$$
 (1)

where RW was the residual weight after ignition.

2.4. Analyses

A representative input sample (0.250 g), samples sieved to different sizes, washed samples and sediments were digested using $HNO₃/HClO₄/HF$. The digestion liquid was diluted and filtered using a 0.22 μ m membrane. The digestion work was carried out in triplicate. The soluble concentration of heavy metals in solution was determined to investigate the extent of heavy metal release during washing. A synthetic precipitation leaching procedure (SPLP) and toxicity characteristic leaching procedure (TCLP) were conducted on the residues to determine the change in toxicity after washing. Cu, Zn, Pb, Ni and Cd in the digestion solution, water and leach liquors were determined by atomic absorption spectrometry (Zeenit 700, Jena, Germany). Energy dispersive spectrometer (EDS) analyses were conducted on the representative sediment samples.

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