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Influence of thermal treatment on fixation rate and leaching behavior of heavy metals in soils from a typical e-waste processing site

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ABSTRACT

A solidification and stabilization method, thermal treatment, was used to investigate the effects of incineration temperature and incineration time on heavy metal fixation rates and leaching behaviors in soils from an e-waste processing site. High concentrations of heavy metals (Ni 153 mg/kg, Cu 448 mg/kg, Zn 227 mg/kg, Cd 0.80 mg/kg, Sn 838 mg/kg, Sb 658 mg/kg and Pb 114 mg/kg) were detected in these soils. In addition, the Mn (67.2%), Co (62.7%), Ni (84.8%), Cu (88.3%), Zn (56.5%), Cd (79.9%) and Pb (67.4%) in these soils mainly occurred in the non-residual fractions. Thermal treatment experiments indicated that better (>80%) Be, V, Cr, Mn, Co, Ni, Cu, Zn, Cd, Sn and Sb fixation rates could be obtained in these soils at an incineration temperature of 700 °C and at an incineration time of 45 min. under these conditions, the concentrations of Be, Cr, Co, Ni, Zn and Cd in the Toxicity Characteristic Leaching Procedure (TCLP) leachates obviously decreased to concentrations that were lower than the corresponding background concentrations of groundwater in Dutch standard, and the Cu concentration in the TCLP leachates decreased from 461 μg/L (before incineration) to 66.4 μg/L. Therefore, thermal treatment technology could be serve as an appropriate measure for Be, Cr, Co, Ni, Cu, Zn and Cd remediation in the soils from the e-waste processing site.

1. Introduction

Heavy metal pollution in soils is of concern due to the rapid industrialization and urbanization that has occurred in developing countries over the past 30 years [10,39]. Heavy metals in soils may result from the weathering of parent materials, and from the accumulation of metals released from human activities [9]. The predominant anthropogenic sources of heavy metals in soils include metalliferous mining and smelting, waste incineration, agrochemicals, organic amendments, electronics, and warfare and military training [13,9,7]. Chromium (Cr), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd) and lead (Pb) are a few of the most common heavy metals in soils [41,38,14,33,12]. Soils can act as a significant sink for heavy metal pollutants in the terrestrial environment [13]. Excessive heavy metal accumulation in soils can degrade soil quality and affect ecosystem functions [23,29]. Furthermore, these metals can be transported from soils to underground water or crops, and negatively affect animals and humans health through food chains (soils–animals–humans) [17]. Therefore, it is important to develop the efficient and suitable technologies for remediating soils that are heavily polluted with heavy metals.

Soil heavy metal remediation technologies primarily include chemical leaching, solidification and stabilization, covering unpolluted soils, drainage systems and dilution etc. [6,2,5,18,19,21]. Chemical leaching may destroy the soil structure and may require the addition of some inorganic acids (e.g., H_2SO_4, HNO_3, HCl), organic acids (e.g., citric acid, malic acid, oxalic acid) or organic chelating agents (e.g., EDDS, EDTA) [2,36], which may lead to secondary pollution. Heavy metals in contaminated soils may be transported to ambient environment through biochemical processes if these soils are covered by unpolluted soils. Drainage system was mainly used in agricultural fields where were slightly polluted by heavy metals [18,19]. However, one of solidification and stabilization method, thermal treatment technology, could change the chemical speciation of heavy metals in soils and reduce their bioavailability [15,24,1,11]. Therefore, thermal treatment may be a suitable remediation technique for soils contaminated with heavy metals, especially for soils polluted with several different metals.

In the past, thermal treatment technology was mainly used for sewage sludge disposal and treatment. Zhang et al. [40] investigated the transfer of heavy metals during sewage sludge

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incineration, and showed that more than 80% of Cr, Cu and Ni, 74–94% of Zn, and 46–79% of Pb remained in bottom ash after incineration. Hyks et al. [11] observed that Cu and Pb leaching could decrease by one order of magnitude after the thermal treatment of municipal solid waste. A survey by Chen and Yan [4] showed that high incineration temperatures and residence times promoted the complete oxidation of metals, reduced the non-residual fractions of metals and decreased the bioavailability of metals.

Preliminary experiments indicated that the soils [27] and sediments [26] in the e-waste acid leaching site from Guiyu were seriously contaminated with Cu, Zn, Cd, Sn, Sb and Pb. A correlation analysis indicated that the metals in these soils were most likely released from uncontrolled e-waste recycling activities [27,26]. It can be inferred that multiple metals contaminate soils and sediments at e-waste recycling sites may be always heavily polluted by multi-metals. However, heavy metal remediation technologies for soils from e-waste recycling sites have rarely been reported, thus, it is important to take effective measures to remediate these contaminated soils.

The objectives of this study were to (1) investigate the influences of thermal treatment on the metal fixation rates in soils from a traditional e-waste recycling site, (2) study the effects of thermal treatment on the leaching behaviors of these soils, and (3) investigate the feasibility of thermal treatment on the remediation of these soils. It is hoped that these findings will provide scientific evidence for remediating soils polluted by various heavy metals in similar areas.

2. Materials and methods

2.1. Sample preparation

Twenty-three soil column samples (80 cm), including 10, 5 and 8 samples from an acid leaching site, a deserted area and a deserted paddy field, respectively, were collected from Guiyu in May 2013. Detailed information regarding the sampling sites can be found in Quan et al. [27]. Each sample was divided into 3 parts: surface soil (0–20 cm), middle soil (30–50 cm) and deep soil (60–80 cm). All 69 subsamples from the 23 soil column samples were placed in polyethylene bags (Ziploc) and stored at 4 °C prior to pretreatment.

All 69 subsamples were thoroughly dried in an oven at 80 °C for at least 2 days, crushed to obtain a fine powder in an agate mortar, passed through a 100-mesh nylon sieve to remove stones, fine debris and dead organisms, and thoroughly mixed and homogenized. One average soil sample, was prepared by mixing equal amounts (≈8 g) of each of the 69 subsamples, and was used for the thermal treatment experiment and Toxicity Characteristic Leaching (not Leachate) Procedure (TCLP) experiment.

2.2. Thermal treatment experiment

The dried average soil samples were incinerated in a muffle furnace. The influences of temperature (400–1000 °C) and time (10–240 min) on the heavy meal fixation rates was determined.

2.3. Sample analysis

2.3.1. Basic physi-chemical characteristics analysis

The pH (water/soil = 2.5:1) of the average soil sample was analyzed using a Leici pH meter (pHS-3C, China). The C, H, N and S contents of the average soil sample were measured using a German elemental analyzer (Vario EL-III). The total organic carbon (TOC) content of the average soil sample was analyzed using a TOC analyzer (CM250, USA) after removing inorganic carbon using 5% (v/v) HCl.

2.3.2. Heavy metal analysis

The average soil sample and incinerated soil samples collected from thermal treatment experiments were digested with a mixture of concentrated acids ($HF/HNO_3/HCl$ = 5:5:2) [13] using a hot plate (ML-2-4, Beijing, China) for the determination of 12 heavy metals at 200 °C. The heavy metals concentrations in these soils were analyzed using an inductively coupled plasma-mass spectrometer (ICP-MS, Agilent 7700X, USA). The instrument was calibrated using internal standards (0.05 mg/L of ^{4}Li, ^{42}Sc, ^{72}Ge, ^{103}Rh, ^{115}In and ^{219}Bi). The detection limits for Be, V, Cr, Mn, Co, Ni, Cu, Zn, Cd, Sn, Sb and Pb were 0.003, 0.006, 0.051, 0.018, 0.009, 0.038, 0.140, 0.263, 0.004, 0.008, 0.003 and 0.006 μg/L, respectively. In addition, replicates of the 10% samples and standard reference materials (GBW07407 and GBW07429) were used or quality assurance and quality control purposes. The recovery rates of the 12 heavy metals in the standard reference materials (GBW07407 and GBW07429) were approximately 92.5–119%.

2.3.3. TCLP leaching experiment

TCLP is widely used to determine the mobility of organic and inorganic pollutants present in liquid, solid and multiphasic wastes. In the United States, TCLP has generally been used as a general method for assessing the toxicity of contaminants in ecosystems [3,31,30]. The detailed leaching procedures of the average soil samples and incinerated soil samples included the following: (1) two different buffered acidic leaching extraction fluids (Fluid 1: CH_3COOH, pH 4.93 ± 0.05 for sample pH < 5 and Fluid 2: CH_3COOH, pH 2.88 ± 0.05 for pH > 5) were prepared for TCLP. In this study, fluid 1 was chosen based on the alkalinity of the average soil sample (pH 2.63 < 5). (2) An accurately weighed soil sample (≈2.00 g) and 40 mL of fluid 1 were transferred into a 100 mL plastic container and rotated in a horizontal shaking mixer for 18 ± 2 h at a speed of 30 ± 2 rpm. (3) The leaching liquid was filtered and passed through a 0.45 μm micron filter membrane. (4) The heavy metal concentrations in the leaching liquid were analyzed using an Agilent 7700X inductively coupled plasma-mass spectrometer (ICP-MS). Reagent blanks and replicates of the 10% samples were also analyzed in this study.

2.4. Data analysis

After thermal treatment, the percentages of the heavy metals in the average soil sample remaining in the slag (fixation rate, R%) were calculated using the following formula:

$$R = \frac{C_2 \times m_2}{C_1 \times m_1} \times 100\% \quad (1)$$

where C_1 is the heavy metal concentration in the average soil sample, mg/kg; C_2 is the heavy metal concentration in the slag after thermal treatment, mg/kg; m_1 is the mass of the average soil sample used in this experiment, g; and m_2 is the mass of slag after thermal treatment of the average soil sample, g.

3. Results and discussion

3.1. Basic physi-chemical characteristics

The basic physical and chemical characteristics of the soil collected from a typical e-waste processing site and its surrounding environment are shown in Table 1. The pH could significantly