Journal of Hazardous Materials 162 (2009) 1000–1006

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Evaluation of the distribution patterns of Pb, Cu and Cd from MSWI fly ash during thermal treatment by sequential extraction procedure

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ARTICLE INFO

Article history:
Received 15 January 2008
Received in revised form 26 May 2008
Accepted 27 May 2008
Available online 7 June 2008

Keywords:
MSWI fly ash
Sequential extraction procedure
Washing pretreatment
Bottom ash
Retention time

ABSTRACT

Municipal solid waste incinerator (MSWI) fly ash was frequently classified as hazardous materials as the metals' concentration of toxicity characteristic leaching procedure (TCLP) exceeded regulations. Many studies have focused on reducing the concentration of TCLP using thermal treatment and increasing the application of thermally treated slag. However, the metal patterns in MSWI fly ash with or without thermal treatment have seldom been addressed. The main objective of this study was evaluation of the distribution patterns of Pb, Cu and Cd from MSWI fly ash during thermal treatment by sequential extraction procedure. The experimental parameters included the form of pretreatment, the proportion of bottom ash (bottom ash/fly ash, B/F = 0, 0.1 and 1) and the retention time. The results indicated that (1) In comparison to raw fly ash, the distribution patterns of Pb, Cu and Cd become stable in thermally treated slag. (2) Washing pretreatment caused the Pb pattern to become stable, while the influence on Cu and Cd were not significant. (3) The distribution patterns of Pb, Cu, and Cd became more stable as the retention time increased. (4) Adding bottom ash could make the distribution patterns of Pb and Cd more stable.

1. Introduction

Because of the recent economic development in Taiwan, a great deal of garbage needs to be appropriately treated and disposed. As Taiwan is a small island country having a high population density, solid waste is primarily dealt with by incineration. The Taiwan government has established approximately 22 municipal solid waste incinerators (MSWI). Incineration has many advantages, such as reducing waste volume and detoxifying waste. However, incineration also generates some secondary pollutants such as gases and particle pollutants. The gases and particle pollutants generated are NO_x and SO_x, and fly ash and bottom ash, respectively. During the combustion process, some metal compounds are vaporized at high temperature and condensed on MSWI fly ash at low temperature in an air pollutant control device. The metals' concentration in toxicity characteristic leaching procedure (TCLP) test leachates of MSWI fly ash, especially for lead, often exceeds the Taiwan EPA regulation. Therefore, MSWI fly ash is considered a hazardous material [1–3]. As a great quantity of MSWI fly ash is produced annually, numerous studies have investigated MSWI fly ash treatments to reduce the metals' concentration in the TCLP test leachates and increase recycling and the reusing potential of MSWI fly ash. Some studies evaluated pollutant emissions during the treatment process [4–10]. In addition to treatment methods for MSWI fly ash, the leachability of heavy metals from treated slag is also a great concern. The sequential extraction procedure developed by Tessier et al. [11] is often used to identify the speciation of heavy metals. This method focuses on the breakdown of materials into varied fractions that can be selectively dissolved by using particular extraction agents. The fractions are often defined as follows:

(I) The exchangeable fraction is easily extracted with solutions in neutral condition and changes in water ionic composition are likely to affect the sorption–desorption processes.
(II) The carbonate fraction is associated with sediment carbonates. This fraction is soluble under acidic conditions and susceptible to changes of pH.
(III) The iron and manganese oxides are excellent scavengers for trace metals and are thermodynamically unstable under anoxic conditions. This fraction can extract iron with a reducing solution followed by an acid or a complex agent.
(IV) Trace metals may be bound to various forms of the organic matter fraction (notable humic and fulvic acids). In order to release these metals, the organic matter can be degraded under oxidizing conditions. Hydrogen peroxide is generally used as extraction agent for such organic matter.

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0304-3894/$ – see front matter © 2008 Published by Elsevier B.V.
doi:10.1016/j.jhazmat.2008.05.155

(V) The residue contains mainly primary and secondary minerals that retain metals in their crystalline structure. These metals in residue are not normally expected to be released into the environment under natural conditions and can be decomposed by digestion with oxidizing acids.

Sequential extraction procedure was applied to solids, sludge, MSWI fly ash and bottom ash. Fuentes et al. [12] applied sequential extraction to different sewage sludges obtained from wastewater treatment plants. Analytical results by Fuentes showed that chromium and copper were most abundant in organic and residual fractions, while zinc and nickel had no dominant chemical phase. Dermatas et al. [13] reported that the percentage of the Pb fraction in soil followed the sequence: carbonate fraction > iron and manganese oxides fraction > organic matter > exchangeable > residual. Over 95% of extracted Pb was associated with carbonate, iron and manganese oxide fractions. Previous studies that investigated the release of metals from MSWI bottom ash determined that almost all Cr and major fractions of Mn were extracted with strong acid. Copper was extracted under oxidizing conditions, and Pb and Zn were acid soluble and easily released into the environment [14,15]. Moreover, some studies have focused on metal fractions in MSWI fly ash using sequential extraction. The experimental results obtained by these showed that most fractions of toxic metals, such as Cu, Cd, Zn, and Pb, accumulated in exchangeable and carbonate fractions. The main fraction of Cd was bound to carbonate and soluble compounds. The chief fractions of Pb, Zn, and Cu were bound to carbonate and organic matter. The physical and chemical properties of MSWI fly ash, such as particle size and composition, also influenced the decomposition of metals obtained by sequential extraction [16–24]. Many studies have focused on metal fractions using the sequential extraction method. However, very few reports have compared metal fractions in raw ash and treated slag. Chou et al. [25] evaluated the emissions of Pb and polycyclic aromatic hydrocarbons (PAHs) from thermally co-treated MSWI fly ash and bottom ash process. In that study, the experimental results only showed the TCLP concentration of Pb. However, the metal fractions in raw fly ash and thermally treated slag with different operational conditions were less to mention.

The aim of this study is to evaluate the patterns distribution of Pb, Cu, and Cd patterns from MSWI fly ash during thermal treatment by sequential extraction procedure. Thermally treated and untreated metal fractions are compared. In addition to thermal treatment, the effects of parameters, such as retention time, pretreatment, and co-treatment on metal fractions are also discussed.

2. Experiment

2.1. Materials

The MSWI fly ash and bottom ash samples used in this study were from a mass-burning incinerator in Taiwan. The MSWI fly ash and bottom ash were sieved to ensure that they contained particles of the same size, respectively. The full chemical and mineralogical characterization of raw fly ash, washed fly ash and bottom ash listed in Table 1 were performed and published in the previous papers [4,26].

2.2. Apparatus

Thermal treatment of the fly ash was conducted in laboratory apparatus and the experiment apparatus of this study was similar to that used by Chou et al. [25]. This apparatus was a pilot-scale rotary kiln reactor with a thermal chamber (210 cm long with an internal diameter of 9 cm), control system and collection system. The thermal chamber was made of steel (AISI 316) and utilized an electrical heater. It had both a temperature control system and a rotary kiln speed controller. The temperature control system had two thermocouples and a proportional integral derivative (PID) controller for temperature adjustment. The rotary kiln speed controller adjusted rotary kiln speed. The collection system was made of steel (AISI 316) and collected the thermally treated slag that fell from the rotary kiln.

Table 1
Main composition of raw fly ash, washed fly ash and bottom ash

Composition (%)	Raw fly ash	Washed fly ash	Bottom ash
CaO	35.81	41.54	18.21
SiO_2	7.62	15.15	24.49
Al_2O_3	3.38	7.29	2.67
Fe_2O_3	1.24	1.92	17.07
MgO	1.41	3.96	1.39
Cl^-	17.74	9.37	NA

Source: [4] and [26]. *Note*: NA, not available.

2.3. Experimental procedure

The experimental procedure had some steps. The MSWI fly ash was washed with distilled waster for 3 h in agitated vessels under room temperature (25 °C). The volume of agitated vessels was 2 L and the agitation rate was 30 rpm. The mass ratio of ash/water was 1:10. After washing, the solid/water mixture was separated via a glass fiber filter. The remaining solids were dried overnight in an oven at 105 °C. This part of the fly ash sample was classified as washed fly ash, and the remaining portion of MSWI fly ash was called raw fly ash. Second, MSWI bottom ash was added to both washed and raw fly ash in various ratios and mixed well. Three ratios of MSWI fly ash (F) to bottom ash (B), B/F = 0, 0.1 and 1, were used. Finally, the ash and mixed ash were fed into the thermal chamber under operational conditions. The operational temperature was 700 °C and the parameters included the form of pretreatment, the proportion of bottom ash (bottom ash/fly ash, B/F = 0, 0.1 and 1) and the three different retention time. The three different rotation rates included 0.89 rpm, 1.39 rpm and 2.0 rpm in this study. The detail operational conditions of this experiment were similar to those used by Chou et al. [25].

2.4. Sequential extraction procedure

The sequential extraction procedure was a modified version of that developed by Tessier et al. [11]. The equipment used for the sequential extraction procedure was 50 mL polyethylene (PE) vessel and into which the sample (2.5 g) was put. The sample and extraction solution were mixed by using a rotation machine and the heating equipment was water bath. All of the extraction conditions were summarized in Table 2. After extracting each fraction, the extraction solution was centrifuged (3000 rpm, 15 min) and filtered. The metal concentrations of each fraction were analyzed by flame atomic absorption spectroscopy (FAAS).

3. Result and discussion

3.1. Metal fraction in raw ash and washed ash

3.1.1. Distribution of Pb pattern

Fig. 1(a) shows the distribution of the Pb pattern in raw ash and washed ash. The main fraction in raw fly ash was carbonate (about 56%). The secondary fraction of raw fly ash was Fe–Mn oxides (about 21%). According to the experimental results, the lead fractions of