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Stabilization of heavy metals in fly ashes from municipal solid waste incineration via wet milling

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ABSTRACT

This study reports the stabilization of toxic heavy metals in fly ashes from incineration of municipal solid waste (MSW) in a grate furnace incinerator (GFI) and a fluidized bed incinerator (FBI) using wet milling without water-extraction pretreatment. After wet milling for 24 h, the relative leaching rates (RLRs) were decreased for all the studied heavy metals (Cr, Cu, Zn, Cd and Pb) and met the regulatory leaching concentrations as determined by Toxicity Characteristic Leaching Procedure (TCLP). For the GFI fly ash, the RLRs of Cr, Cu, Zn and Pb decreased from 11.00%, 4.27%, 5.12%, and 72.64% to 0.81%, 0.08%, 0.33%, and 1.12%, respectively. The decreased rate of RLRs for all four heavy metals had exceeded 90%. For the FBI fly ash, the RLRs of Cr, Cu, Zn, Cd and Pb decreased from 6.81%, 0.09%, 1.99%, 8.92%, and 0.10% to 1.51%, 0.05%, 1.14%, 3.05% and 0.02%, respectively. European Community Bureau of Reference (BCR) sequential extraction results demonstrate that, for the GFI fly ash, wet milling led to decreases in the water- and acid-soluble and reducible fractions, but an increase in the more stable residual fraction. For the FBI fly ash, except for Cu, the residual fraction increased while the water- and acid-soluble fraction largely decreased after wet milling.

1. Introduction

Municipal solid waste incineration (MSWI) is considered as an effective approach for energy recovery and pollution minimization in China [1–3]. However, a large quantity of fly ash is produced from MSWI, with an annual production rate of over 200,000 tons [4], which is expected to increase due to increased numbers of waste incinerators. Currently, fly ash from MSWI is mostly landfilled [5,6], which poses serious environmental issues because the ash is classified as hazardous solid waste and enriched with leachable toxic heavy metals, such as Cu, Cd, Cr, Zn and Pb [7–9]. Therefore, MSWI fly ash must be pretreated to minimize its environmental impacts upon landfilling.

Chemical stabilization is one of the most economically feasible pretreatment methods because of its lower capital investment [10]. This method stabilizes the heavy metals in MSWI fly ash via altering their occurrence forms from soluble fractions to less soluble ones [11], reducing their leachability and thereby improving their stability [12,13]. The stabilization of heavy metals can also be achieved by wet milling treatment, which can help the heavy metals exposed in the inner layer and enhanced the reactivity and thereby change the occurrence forms of heavy metals [14].

Wet milling treatment has been used to stabilize heavy metals [14–21], particularly lead (Pb), in MSWI fly ash. This treatment is considered as an environmentally friendly approach that can make the particle size smaller and activate the reagent [22,23]. For example, Chen et al. [15] used this method for treating MSWI fly ash that was then added to an ordinary Portland cement. They found that the milling process helped stabilize the heavy metals in the MSWI fly ash and had an activating effect after a water extraction process. Their results demonstrated that, except for Cr, all other heavy metals investigated were stabilized in the milled sample because of the concurrent fragmentation and/or agglomeration. Sun et al. [17] found that wet milling treatment can convert Pb from a readily soluble fraction to a hard-to-leach fraction. They discovered that the treatment efficiency for Pb increased from −55.18% to over 40% with the milling time increasing from 1 to 24 h when the milling solution is water. Li et al. [18] noted that 93.11% Pb was partitioned from ash using desalinated water as the milling water when the MSWI fly ash was water-extracted. The studies above confirm the critical role of wet milling treatment in stabilizing heavy metals in fly ashes produced by grate-furnace incinerators (GFIs). However, it is questionable whether wet milling is applicable in stabilizing heavy metals in fly ashes from fluidized-bed incinerators (FBIs),

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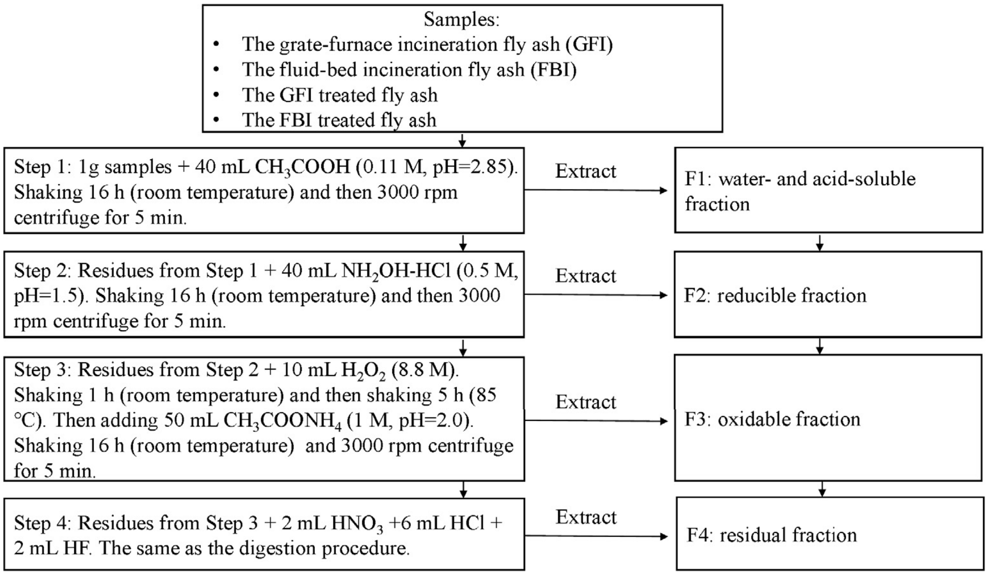

Fig. 1. Detailed information of BCR procedure.

which are another primary type of incinerator commonly used in China [3,24]. In FBIs, coal is generally added into the MSW, the fly ash of which is enriched with certain toxic elements such as Pb, Cr, Cu, Zn, and Cd [25]. Additionally, previous studies have employed water extraction before wet milling [15,17,18]. Thus, it is desirable to investigate whether water extraction and wet milling treatment can be integrated into one step. Furthermore, the transformation of trace elements in MSWI ashes after wet milling treatment remains unclear.

This study aims to systematically investigate the technical feasibility of stabilizing five typical toxic trace elements (i.e., Pb, Cr, Cu, Zn, and Cd) using wet milling treatment for various times ranging from 1 to 24 h in two types of MSWI fly ashes, one produced from a GFI and the other from a FBI. European Community Bureau of Reference (BCR) sequential extraction procedure was conducted for both the raw and treated fly ashes to reveal the effects of wet milling treatment on the speciation of the five trace elements.

2. Experimental section

2.1. Samples

The MSWI fly ashes were collected from a GFI and a FBI in Wuhan, Hubei Province, China. Both samples were collected from the hopper of the bag house filters. The two samples were homogenized, dried at 105 °C for 24 h, and sieved to 40–70 μm, prior to further experiments and analyses, since ~90% of the fly ashes are within this size fraction.

2.2. Wet milling treatment

The wet milling treatment of the two fly ashes was conducted using a planetary ball mill (Model: QM-3SP2). For a typical experiment, ~8 g of fly ash and 80 mL of ultrapure water were charged into a 250 mL zirconia vial of the planetary carrier, along with 30 and 22 zirconia balls with diameters of 6 and 20 mm, respectively. The weight ratio of the balls to the ash was ~12, identical to that used in a prior study [26]. The wet milling treatment was conducted for various times (1, 6, 12, and 24 h) at a rotation speed of 250 rpm, which was calculated according to the equation proposed by Rose and Sullivan [27]. After milling, the ash slurry was discharged into a glass beaker. All the milling balls and the planetary vial were repeatedly washed with ultrapure water for 3 times to remove any heavy metals adhered, the liquid from which was also discharged into the glass beaker. The collected ash slurry was then filtrated to separate the treated ash from milling solution. Typical heavy metals, including Cr, Cu, Zn, Cd and Pb, were studied because of their severe toxicity and relatively high concentrations in the fly ashes. Since the wet milling process is accompanied by water leaching, to evaluate the role of wet milling treatment, the two fly ashes were also leached with ultrapure water under conditions identical to those used for wet milling, without using the zirconia balls.

2.3. Leaching of the raw and wet milling treated fly ashes

Two types of leaching experiments were conducted for the raw and treated fly ashes. The first type followed the procedure outlined in the Toxicity Characteristic Leaching Procedure (TCLP) Standard (EPA 1311) [28]. For a typical leaching experiment, ~1.5 g of fly ash was added to ~30 mL of acetic acid solution (pH value of 2.88) at a liquid/solid ratio of 20 mL/g. After blending for 18 h, the mixture was filtered and the supernatant was analyzed for the concentrations of the five trace elements. Here, we defined a leaching indicator, the relative leaching rate (RLR) in%, as calculated by Eq. (1):

$$RLR = CV/(C_0 m_0) \times 100\% \quad (1)$$

where C is the mass concentration of certain trace element in the supernatant, mg/L; V is the volume of leaching solution, mL; m_0 is the mass of fly ash used in the TCLP procedure, g; and C_0 is the concentration of the trace element in the raw or treated fly ash, mg/kg.

The second type of leaching experiment was performed following the BCR procedure to determine the occurrence forms of the trace elements in the fly ashes before and after wet milling. A typical BCR leaching experiment consists of four extraction steps [11], resulting in the corresponding fractions of F1, F2, F3 and F4 (see Fig. 1). The fractions of F1 and F2 were more leachable, while those of F3 and F4 were more stable, particularly F4, which was considered as not leachable in the environment. These leaching experiments were repeated at least three times to ensure reproducibility.

2.4. Sample analysis

The chemical compositions of the raw and treated fly ashes were analyzed using X-ray fluorescence (XRF) spectroscopy (model: EAGLE III). The primary mineral phases in these samples were identified using X-ray diffraction (XRD, model: X'pert PRO) with a 2θ angle ranging from 0 to 60°. A scanning electron microscope (SEM, model: JEOL-7800F) was also used to observe the effects of the wet milling process on the morphology of the fly ashes. The Brunauer–Emmett–Teller (BET)