



Research article

Effects of pH dynamics on solidification/stabilization of municipal solid waste incineration fly ash

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ABSTRACT

Fly ash (FA), a product of municipal solid waste incineration (MSWI), has been classified as a kind of hazardous waste due to its high content of heavy metals. FA may be reused in the construction industry or disposed of at landfill sites, and thus poses threats to both the environment and human health. This study sought to establish a scientific basis for accurate selection of suitable pH storage conditions for the FA. We evaluated the potential of MSWI FA sample from the Xinghuo waste incineration power plant, Wuhan, to solidify/stabilize the heavy metal (Cu, Pb, Zn, Cr, Cd, As and Mn) contents, when leached under different pH conditions. The concentration of a heavy metal in the leachate was assumed to inversely reflect the extent of its solidification/stabilization (S/S). The study findings showed that the raw FA contained higher levels of the heavy metals, which were above the acceptable limits. Extremely acidic conditions favoured heavy metal leaching compared to extremely alkaline conditions. The extent of S/S of heavy metals was generally very low under highly acidic conditions ($\text{pH} \leq 4$), but increased with increasing pH. All the metals solidified/stabilized in pH media of 5–11, except Zn which was detected in the entire pH range. We conclude that changing landfill conditions which can affect the pH environment, will increase heavy metal leaching when the $\text{pH} \leq 4$. As a result, waste which was initially classified as non-hazardous may later pose harmful risks to both humans and the environment alike. We propose pH of 5–11 as the optimum pH range for the treatment, reuse, and disposal of the ash sample.

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1. Introduction

Municipal solid waste (MSW) incineration has become necessary due to the increase in waste generation rates caused by population increase and advancement in the socio-economic wellbeing of both developed and developing economies. The increase in MSW generation has put pressure on city authorities who are faced with limited land space and the challenge to find alternative treatment methods. Also, this upsurge in MSW generation has brought its own opportunities such as providing raw materials for the waste-to-energy (WtE) sector.

According to Lam et al. (2010) incineration of MSW can reduce the waste volume and mass by up to 90% and 80%, respectively. This process of waste treatment is referred to as waste-to-energy if

energy is recovered during incineration. Usually, the heat generated from waste is used to produce steam in boilers for energy production. Some facilities also use the same incineration process to provide heating for public and industrial usage (Palanivel and Sulaiman, 2014; Scarlet et al., 2015). In most instances where cities cannot afford incineration facilities capable of energy recovery and other uses, incineration is done to only achieve reduction in quantities. It may be preferred in cities where space for landfill construction and operation is scarce.

The waste incineration technique produces secondary waste in the form of ashes broadly known as bottom ash (BA) and fly ash (FA), which have to be landfilled or reused. The BA is classified as non-hazardous, and as a result can be landfilled with little or no treatment (Li et al., 2014; Pan et al., 2008). Fly ash however, is listed as a hazardous material due to the presence of deleterious substances that require special treatment and disposal (Huang et al., 2011; Jiao et al., 2016; Xiong et al., 2014; Zhou et al., 2016). Though incineration is able to reduce the quantity of MSW, it potentially poses risk to both the environment and human health if

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its resulting ashes are not properly handled and effective controls are not installed within the incineration facility (Chang et al., 2009; Lam et al., 2010; Li et al., 2016; Sun et al., 2016).

Management options for FA include treatment and disposal in monofills or co-disposal with bottom ash. This may be preceded by separation processes aimed at reducing the chloride, salts, alkali and heavy metals. As it has been suggested by Sun et al. (2016), separation processes reduce the toxicity of the ash, thereby making it suitable for reuse in asphalts, cement, concrete and landfill cover. Heat treatment is also employed to effectively vaporize heavy metals in FA (Yu et al., 2015).

Solidification/Stabilization (S/S) treatment technology for FA is one of the internationally accepted treatment criteria. Chang et al. (2009) defined solidification as the process of converting FA into a monolithic form or granular material, thereby making it relatively easy to handle and haul to final disposal sites. On the other hand, stabilization involves the conversion of the FA to a physically and chemically stable form (Chen et al., 2009; Saeed, 2012; Singh and Pant, 2006). According to Sun et al. (2016) the S/S process is widely used because large proportions of FA is still sent to the landfill. As a result, it is used to treat hazardous waste in order to reduce its toxicity and facilitate other processes prior to landfilling (Luna Galiano et al., 2011; Tang et al., 2016). This treatment method involves adding a reagent into the contaminated media or waste. Binding material commonly used for S/S include reagents, portland cement, lime, cement kiln dust, lime, kiln dust, fly ash, limestone slag, gypsum, and phosphate mixtures (Malviya and Chaudhary, 2006; Wilk, 2007). Owing to the great differences of waste components and media, a blended design ought to be carried out on each waste category. Most S/S binders are made of a mixture of the aforementioned inorganic reagents.

Regulated leaching test criteria used to assess the leaching of FA include the UNE EN 12457-4 and USEPA TCLP. The UNE EN 12457-4 test is an extraction method similar to that of TCLP, but the extraction medium is distilled water, in an L/S ratio of 10 for 24 h (Baun et al., 2003). The USEPA TCLP-1311 method consists of stirring granular material (<9 mm), and using acetic or other prescribed solutions with an L/S ratio of 20 for 18 h (USEPA, 2011).

Several effective media have also been developed by a number of researchers to digest and extract the contents of fly ashes prior to the determination of their heavy metal levels. These include the use of CH₃COONa in batch leaching experiments (Liu et al., 2005), application of CH₃COOH/NaOH as an extracting medium (Singh and Pant, 2006), the use of de-ionized water and HNO₃ solution for extraction (Li et al., 2007), digestion with water followed by HCl extraction (Huang et al., 2011), and using HNO₃/NaOH as leachates (Billen et al., 2014). However, we came across a limited number of papers published to date that have reported on the extraction of heavy metals from MSWI FA using HCl. Huang et al. (2011) focused on the comparison of the leaching effect of acids such as HCl, C₆H₈O₇, C₄H₆O₅, CH₃COOH, CH₃CH(OH)COOH, C₂H₆O₆, C₄H₆O₆, H₂SO₄, and HNO₃ using specific pH values of 3.10, 3.13, 3.07, 3.02, 3.06, 3.09, 2.98, 3.03, and 3.01, respectively. Liu et al. (2005) focused on pH of 2, 4, 6, 10, and 12, while Zhang et al. (2016) used pH of 2, 4, 5.5, 7, 8, 9, 10.5, 12, and 14. However, this study broadened the scope of investigations on the application of HCl in the FA heavy metal leaching tests.

This study sought to present scientific based evidence of the possible effects of the treatment and disposal of FA on the environment and human health. We researched to determine an environmentally safe pH range for treatment and disposal of municipal solid waste incineration (MSWI) FA. A novel approach was adopted where (1) the entire range on the pH scale was considered, and (2) the pH of the various media was adjusted with HCl or NaOH, as an attempt to develop a new leaching procedure

for FA. This is to help eliminate possible short to long term risks that may result from the current limited information and data in terms of pH on the leaching characteristics of FA, which have implications on treatment and disposal procedures and practices. Moreover, much environmental research is needed to guide policy development on sustainable treatment of FA. This study will open up the scientific discourse on FA which is likely to avail basic as well as applied research opportunities in the era of increasing MSWI. Furthermore, based on the literature we came across, this research is among the few studies done pertaining to the S/S potential of MSWI FA, coupled with the possible pH fluctuation impacts on its leachability under mimicked conditions using pH values ranging from 1 to 14. The use of HCl other than the conventional HNO₃, which resulted in similar results of other pH-related studies serves as an addition to the list of possible acids that can be used by researchers and practitioners in this field. This study further provides a pH guide for the handling, transportation and reuse of FA in order to reduce and/or eliminate health and environmental risk associated with heavy metal leaching.

2. Materials and methods

The FA sample was obtained from Xinghuo waste incineration power plant in Wuhan City, Hubei Province, P. R. China. This facility is designed to handle 1000 tons of household waste per day, which accounts for about 25% of the total domestic waste generated in Wuhan City. The total installed capacity of this plant is 24 megawatts. The technology of Xinghuo waste incineration power plant is the grate furnace process. Incineration of MSW discharges lots of dust and harmful acidic smoke which need to be purified. Gas purification technology of the incineration plant is done by initially injecting hydrated lime (Ca(OH)₂) to neutralize the harmful acid gas, before it is channelled through bag filters to intercept the dust, acid gas neutralization products, residual hydrated lime and so on. The gas purification process produces large amounts of FA, which has harmful effects on humans and the environment due to the presence of heavy metal pollutants. Thus, treatment of the fly ash with respect to heavy metals before disposal or reuse is important.

2.1. Sample preparation for testing

2.1.1. XRF and XRD analyses

The FA sample (particle size < 1 mm) was dried at 105 °C for 24 h before laboratory tests was conducted. The chemical composition of the FA was determined by X-ray fluorescence (XRF, AXIOSmAX, PANalytical B. V.), while the mineral phases were determined by the X-ray powder diffraction (XRD, D8-FOCUS, Bruker AXS). Table 1 and Fig. 1 present the results of the XRF analysis and XRD analysis, respectively.

2.1.2. Determination of heavy metal content and concentration in FA sample

To determine the types of heavy metal and their respective mass concentrations, the FA sample was dried as supplied at 105 °C, and analysed by inductively coupled plasma optical emission spectrometry (ICP-OES). The heavy metals identified in the FA sample were Arsenic (As), Copper (Cu), Lead (Pb), Cadmium (Cd), Chromium (Cr) and Manganese (Mn) (Table 2). Table 2 presents the total heavy metals concentration in the FA sample. In order to classify the present FA sample, its heavy metal content was compared with the USEPA-1311 TCLP standard. The levels of all heavy metals exceeded the USEPA-1311 TCLP standard limits.

2.1.3. Leaching test

To evaluate the S/S of the sampled FA under room temperature,

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