Waste Management 80 (2018) 101–111

Contents lists available at ScienceDirect

Waste Management

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

Characterization of naturally aged cement-solidified MSWI fly ash

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ARTICLE INFO

Article history:
Received 17 June 2018
Revised 30 August 2018
Accepted 31 August 2018

Keywords:
Municipal solid waste incineration fly ash (MSWI-FA)
Solidification
PH-dependent
Geochemical model
Long-term stability

ABSTRACT

Solidification/stabilization (S/S) is the most common treatment for municipal solid waste incineration fly ash (MSWI-FA), and is widely applied in developed countries but has a history barely longer than 10 years in China. However, our understanding of the physicochemical characteristics of the solidified FA body after long-term natural aging is comparatively poor. Focusing on cement-solidified FA that was naturally aged for 6 years (hereafter referred to as FA-6), the physicochemical characteristics including elemental composition, mineral composition, microstructure, thermogravimetry, distribution of heavy metals in mineral phases, and leaching characteristics of inorganic salts (Na, K, Ca), anions (Cl and SO_4) and heavy metals (Cd, Cr, Cu, Pb, Zn) were investigated in this study. By combining pH-dependent leaching results with the geochemical model LeachXS, the chemical forms of heavy metals in the FA solid phase was determined. The main conclusion was as follows: (1) soluble salts of FA-6 decreased by more than 92% compared with fresh FA. (2) In FA-6, the proportions of Pb, Cd and Zn in the non-mineral phase were 100%, 100% and 58%, respectively, which may cause potential environmental risk of heavy metal release. The leaching concentration of Pb was 4007.37 μg/L according to compliance batch test of HJ300, which was far higher than the landfill requirement of 250 μg/L. (3) The controlling phase for Pb in FA-6 was $Pb_5(PO_4)_3Cl$ (pH 2–12) and $Pb_2(OH)_3Cl$ (pH > 12). (4) Carbonates, hydrous Fe oxides (HFO) and dissolved organic carbon (DOC) in FA-6 also affected the phase-controlled leaching of heavy metals. The carbonate fraction partly controlled the leaching of Cd, Cu and Zn. For example, smithsonite ($ZnCO_3$) controlled the release of Zn (pH 2–13). Adsorption to solid humic acid (SHA) controlled the Cr leaching at pH < 7 and the Cu leaching except pH > 12.

1. Introduction

Due to its capacity to significantly reduce the volume, mass and harmfulness of municipal solid waste (MSW), incineration has become the most common method of treating MSW in large and medium-sized cities in China. Currently, about 40% of MSW in Chinese metropolitan areas is incinerated, with the rate increasing annually by 8–10% such that it will reach 60% by 2020 (Wang et al., 2018; Xin-gang et al., 2016). MSW incineration fly ash (FA) is generated from air pollution control systems for flue gas. With the continuous improvement of air pollution controls, the FA collected in such pollution control systems has become more highly contaminated. The basic characteristics of FA in China ae as follows: (1) FA is generated in enormous and growing quantities, due to the increasing amounts of MSW incinerated. In 2016, the generation of FA in China exceeded 5 million tons, and this amount is expected to grow rapidly over the next few years, likely exceeding 7 million tons by 2020 (Wang et al., 2018). (2) Pollutants such as heavy metals and dioxin are concentrated in FA, such that FA is considered hazardous waste in China (Zhang et al., 2016). (3) Large quantities of plastic and food waste in MSW enter the incinerator, resulting in high Cl and soluble salt contents in Chinese FA (Liu et al., 2017a; Liu et al., 2017b).

S/S use additives or binders in order to immobilize physically and/or chemically hazardous components initially present in waste (Wiles, 1996). Six waste types contaminated with heavy metals can be handled by S/S: fines grained soil (clay and sites), coarse grained soils, metal sludges, nitrate salts from processing operations, chlorides salts from off-gas treatment, and incinerator fly and bottom ash (Conner and Hoeffner, 1998). Recently, a novel technique of cold bonding pelletization of waste is employed in S/S for waste reutilization in low cost building materials production. Particularly, artificial aggregates are one of the most interesting technological solutions for waste recovery. Colangelo (2015) achieved a lightweight porous aggregates which are mostly suitable for recovery in the field of building materials with enhanced

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https://doi.org/10.1016/j.wasman.2018.08.053
0956-053X/

sustainability properties. Besides, S/S of FA through a polycondensation reaction to create geopolymers can also be suitable for non-structural applications, such as backfilling of abandoned quarries, brick fireplaces, hearths, et al (Ferone, 2013).

Although FA still landfilled, in some cases, FA are placed in salt mines where they are used as backfill, it would be better to conduct practical applications than merely dump the material in a landfill. Ferreira et al. (2003) concluded a nine potential applications into four main categories: construction materials (cement, concrete, glass); geotechnical application (road pavement, embankments); agriculture (soil amendment), and miscellaneous (sorbent, sludge conditioning). To date, the classification is more inclined to circular economy. Quina et al. (2018) summarized the six main routes to manage FA, two hampering further recovery (backfilling and treatment + landfilling) and four methods with potential for recovery of raw materials or manufacture of products (detoxification; product manufacturing; practical application and recovery of materials). For example, as FA is generally composed of oxides, such as SiO_2, Al_2O_3, CaO and Fe_2O_3, it received attention also as a low cost material for the manufacture of cement production (Yin et al., 2018). However, 5% mixing ratio of FA would significantly increase the overall trace metals from their original concentrations inside cement.

Although many a technological approach of FA has been conducted, "FA S/S + landfill" is still the major application and this technical approach will prevail in next decade. Here we made a serious consideration on the performances of natural aging solidified FA, not involving other techniques. FA cement solidification exploits the cement hydration reaction to produce a high-strength solidified body for physical encapsulation and chemical stability of heavy metals. Cement solidification of hazardous waste has evolved gradually since the 1990s. In European countries, the content of soluble salts in FA, such as chloride and sulfate, is low (2.9–11.5% and 8.0–10.5%, respectively) (Colangelo et al., 2012; Huber et al., 2018; Nowak et al., 2010; Polettini et al., 2001), and thus cement is highly effective for immobilization of heavy metals in FA. In China, because of its advantages of low-cost, speed and efficiency, cement solidification has become the most commonly used pretreatment before landfilling of FA. However, the high content of chlorides can degrade the performance of cement hydration. As a result, the setting time of cement may be longer and its compressive strength reduced, decreasing the immobilization effect on heavy metals (Aubert et al., 2004; Malviya and Chaudhary, 2006a; Quina et al., 2008). Generally speaking, due to limits at the economic and management levels, FA control technology in China is relatively poorly developed. Although cement solidification has been in practice in China for more than 10 years, FA managerial experience is lacking. One of the main reasons for this is due to a scarcity of studies characterizing the natural aging process of solidified FA. Currently, our understanding of the characteristics of solidified FA is limited to the short-term curing stage. However, the characteristics and long-term stability of heavy metals in solidified FA, over and above natural aging, are worthy of exploration.

We analyzed the main physicochemical characteristics of cement-solidified FA after 6 years of natural aging (hereinafter referred to as FA-6), in particular the major and minor mineral phases and leaching characteristics, to determine the leaching level and behavior of heavy metals. We incorporated the results of elemental composition and pH-dependent leaching tests of FA-6 into a geochemical speciation model to determine the major mechanism controlling the release of heavy metals according to pH and geochemical conditions. From the perspective of environmental risk, this paper provides ideas for FA management and landfill treatment and disposal processes.

2. Experimental methods

2.1. Experimental materials

The rapid development of MSW incineration started in the period of 2006–2010, which the number plant increased from 60 to 105 in China. The massive treatment of FA was also started during this period. Specifically, the history of FA cement solidification in this incineration plant was only 6–7 years. Thus the availability of natural aged solidified FA was 6 years old. FA-6 was sampled from a landfill in Jiangsu province. The incineration plant can incinerate the MSW of 1200 t/d. The generation of FA is about 36 t/d. The FA landfilling began at 2009. The solidified FA was landfilled first at 2011. It was generated from a mechanical grate waste incinerator and a flue gas purification process using selective non-catalytic reduction (SNCR), a semi-dry process (lime slurry), and activated carbon. The FA in this study should be called air pollution control (APC) residues. However, the APC residues is called "fly ash" in practice. To enhances the audience, we defined the APC residues as FA.

PO 32.5 cement created during the solidification process comprised 20% FA. The location of the landfill is shown in Fig. 1. The area of sampling was about 20 m * 20 m. The area was far from road and well covered by HDPE film to prevent the disturbances generated by human activities. About 0.5 kg samples at a depth of 15–20 cm from each grid sampling of 4 m * 4m was mixed together and kept in a self-closing plastic bag. Wooden shovel was used to collect the fly ash samples to avoid the heavy metal contamination from the metal tools.

Analysis of core samples of cement-solidified FA obtained at a depth of 1–12 m showed that the leached amounts of certain core components (including high concentrations of soluble salts) did not change obviously with depth and carbonation did not occur (A. et al., 2011; Kosson et al., 2014b). The surface layer of solidified FA is eroded by the natural environment, and thus solidified FA was sampled at a depth of 15–20 cm. Sample collection and preparation were based on the technical standard for sampling and sample preparation of industry solid waste (HJ/T 20-1998).

2.2. Tests of physicochemical characteristics

2.2.1. Elemental analysis

X-ray fluorescence The elemental composition of oxides from the FA-6 sample was analyzed using an X-ray fluorescence spectrometer (XRF). The samples were dried at 105 °C for 30 min,

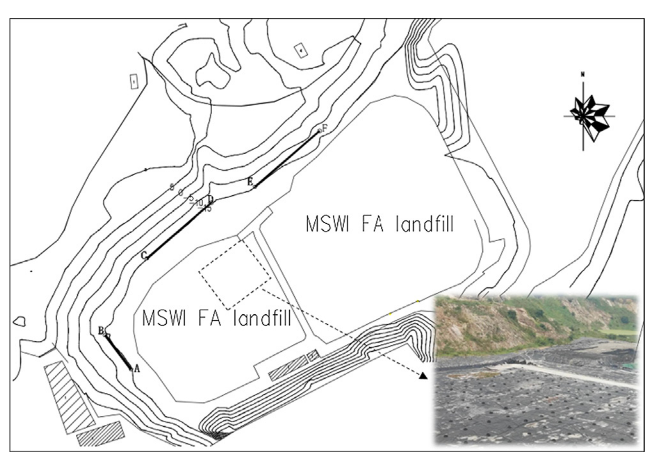

Fig. 1. The location of the FA landfill.