Waste Management 56 (2016) 238–245

Contents lists available at ScienceDirect

Waste Management

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

Stabilization/solidification of municipal solid waste incineration fly ash via co-sintering with waste-derived vitrified amorphous slag

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ARTICLE INFO

Article history:
Received 16 March 2016
Revised 6 June 2016
Accepted 2 July 2016
Available online 16 July 2016

Keywords:
MSWI fly ash
Heavy metals
Chlorides
Sintering
Stabilization/solidification

ABSTRACT

Municipal solid waste incineration (MSWI) fly ash (FA) is classified as hazardous waste and requires special treatment prior to landfilling due to its high levels of alkali chlorides and heavy metals. In this paper we presented and discussed a novel method of converting FA into an inert and non-hazardous material, by using the metastable state of vitrified amorphous slag (VAS). XRD results showed that VAS remained in the amorphous state when sintered at 700 and 800 °C and were in the crystalline state at 900 and 1000 °C. Heavy metals- and Cl-containing phases appeared during phase transformation process. The residual rates of heavy metals and Cl increased with the decrease of FA:VAS ratios. The prolonged leaching test and potential ecological risk assessment of heavy metals showed that the heavy metals were well immobilized into the sintered samples and presented no immediate threat to the environment. The results indicated that the immobilization of heavy metals was due to the reaction with silicate or aluminosilicate matrices within VAS and/or the incorporation into the new generated crystals. The proposed method can be considered as a potential promising technique for the stabilization/solidification of MSWI fly ash with high Cl content.

1. Introduction

Incineration is one of the most common municipal waste management methods and has gone through major development in recent years due to its superiority in mass and volume reduction of waste and energy recover. It has become the dominant approach to the treatment of municipal solid wastes in some developed countries, such as Japan, Sweden and Denmark, and there are now more than 300 MSWI plants in China (Hu et al., 2012; Pan et al., 2013). With the increasing number of MSWI plants, the amount of fly ash has been increasing greatly each year (Gao et al., 2008; Wang et al., 2010; Wu et al., 2011). As an unavoidable byproduct, MSWI FA has been classified as hazardous waste and need special disposal due to various organic compounds, toxic heavy metals, and soluble chlorides (Cyr et al., 2012; Ginés et al., 2009; Li et al., 2012; Shi et al., 2009). The toxic heavy metals can easily leach out and pollute soil and ground-water, posing high potential risks to both the environment and human health.

Various technological options have been reported for the processing of MSWI FA, including cement or geopolymeric solidification (Colangelo et al., 2012, 2015; Cyr et al., 2012; Guo et al., 2014; Su et al., 2016), chemical stabilization using additives (Chen et al., 2013; Sukandar et al., 2009), thermal treatment via melting (Jung and Osako, 2009; Yang et al., 2009), and hydrothermal techniques (Hu et al., 2012; Shan et al., 2012). At present, the main approach to the disposal of MSWI FA in China is to transport to specific landfill sites after stabilization/solidification with cement. The toxic components can be incorporated into the cement matrix through either physical or chemical immobilization. However, MSWI FA has high Cl content due to the polyvinyl chloride in plastics and sodium chloride in the Chinese diet, ranging between 10.8% and 19.6% (Hu et al., 2012; Li et al., 2014; Zhang et al., 2011; Zheng et al., 2011). Chloride is considered to be one obstructive component and will interfere with the basic hydration reactions of cement, leading to an inadequate degradation of the waste form over time (Chen et al., 2013; Cyr et al., 2012). Fortunately, the chloride species, NaCl, KCl, and $CaCl_2$, in MSWI FA are mainly readily water-extractable. For this reason, the water washing pre-treatment is generally carried out as a cheap but effective method to remove soluble chlorides and amphoteric heavy metals from fly ash with proper setting of liquid-to-solid ratio and washing time (Chen et al., 2012; Chiang and Hu, 2010; Ferone et al., 2013b; Yang et al., 2012; Zheng et al., 2011). The resulting washed FA can be further used as secondary raw materials to produce end-use products, such as glass-ceramics, road base, cement and brick (Chu et al., 2013; Galiano et al., 2011; Vu et al., 2012). However, the treatment costs of toxic wastewaters increase with flow rate

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http://dx.doi.org/10.1016/j.wasman.2016.07.002
0956-053X/

even if certain processes (membrane purification, adsorption, and chemical precipitation, etc.) have already treated the water. Therefore, it is necessary to find new solutions to safely dispose of or recycle the quickly increasing amount of MSWI FA.

In recent years, several studies have reported the recycling of fly ash as building materials by thermal processing technique (Bernardo et al., 2009; Chu et al., 2013; Karamanov et al., 2003; Yang et al., 2009) and cold bonding technique (Ferone et al., 2013a; Güneyisi et al., 2015a, 2015b). In our previous research (Zhang et al., 2015), a vitrified amorphous slag (VAS), synthetized by melting oil shale fly ash (OSFA) and MSWI bottom ash (BA), has been used as raw material to prepare glass-ceramics. High temperature melting technique has been identified as one of the most promising methods to safely handle hazardous wastes from the viewpoint of product safety and technical advancement. The majority of heavy metals are evaporated or concentrated in molten slag, which can further synthesize glass-ceramics materials by devitrifying the obtained glass-like slag by following single or two-stage heat treatment. The derivative glass-ceramics material can show good performances and the heavy metals can be well immobilized into the crystal structures. In addition, previous studies have reported the possibility of converting hazardous wastes into safe products by sintering with precursors (Tang et al., 2011, 2013), in which the heavy metals can form phases that can exhibit sufficient stability due to irreversible phase transformation. Therefore, it is possible that MSWI FA could be stabilized by reacting with the metastable VAS due to its large specific surface area and high internal energy.

Based on the analysis above, a novel method was proposed for the stabilization/solidification of MSWI FA with high Cl content by sintering with VAS. In order to explore the feasibility of the proposed method, the present study has been conducted to (i) observe the phase transformation of MSWI FA with different amounts of VAS at temperatures ranging from 700 to 1000 °C for 3 h; (ii) analyze the residual rates of heavy metals and Cl affected by FA:VAS ratio and temperature in the sintered samples quantitatively; (iii) assess the leaching behavior and potential ecological risk of heavy metals from the sintered samples. This study provided some fundamental theories and can be viewed as a basic understanding for new technique of stabilization/solidification of MSWI FA with high Cl content.

2. Materials and methods

2.1. Raw materials

FA and BA were collected from a municipal solid waste incinerator (with a capacity of 1500 ton/d) located in Liaoning province, China. OSFA was supplied by a thermal power plant located in Jilin province, China. The samples were dried at 105 °C, and then graded to pass sieve NO. 150 (the diameter of mesh is 106 μm) for subsequent experiments. Chemical constituents and heavy metals concentrations of FA, BA, and OSFA were determined by X-ray fluorescence (XRF) spectrometry (PDA-5500II, Shimadzu, Japan) and inductively coupled plasma optical emission spectrometry (ICP-OES) (Optima 2000DV, PerkinElmer, USA), respectively. Based on our previous research work, a mixture of 70% OSFA and 30% BA by weight was homogenized in a ball mill for 1 h and used as raw material to synthesize VAS.

2.2. Synthesis of waste-derived vitrified amorphous slag

The mixture of BA and OSFA was heated at a rate of 10 °C/min from 25 to 1500 °C for a duration of 2 h in a high temperature furnace (T_{max}: 1600 °C). The melt was rapidly poured into water, and then dried, ground and sieved into powder (<106 μm) to synthetize

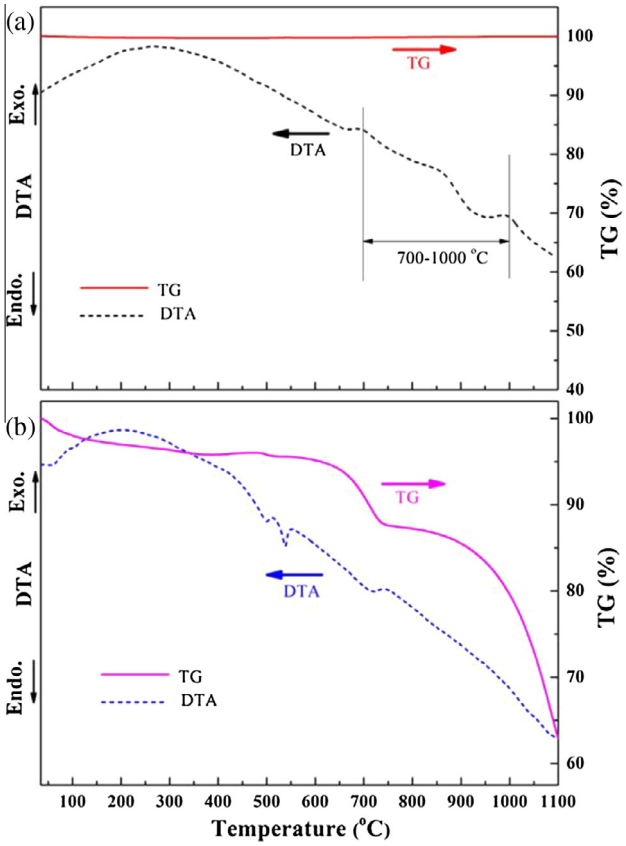

Fig. 1. TG/DTA curves of (a) VAS and (b) FA heating at 25–1100 °C in air at a rate of 10 °C/min.

Table 1
The major and trace element compositions (±Standard Deviations (SD); n = 3) of FA, BA and OSFA.

	Major compositions (wt%)				Trace elements (μg/g)		
	FA	BA	OSFA		FA	BA	OSFA
SiO_2	3.36 ± 0.24	14.28 ± 0.37	55.35 ± 0.25	Cr	549 ± 12	296 ± 14	381 ± 20
CaO	33.60 ± 1.60	55.44 ± 1.04	7.98 ± 0.12	Cu	924 ± 46	458 ± 33	1968 ± 59
Al_2O_3	1.08 ± 0.12	3.68 ± 1.12	17.25 ± 0.65	Zn	7332 ± 219	2631 ± 32	195 ± 15
Fe_2O_3	1.88 ± 0.20	4.87 ± 0.49	10.45 ± 0.65	Pb	2703 ± 81	334 ± 8	59 ± 7
Na_2O	9.22 ± 0.77	1.26 ± 0.20	0.91 ± 0.03				
SO_3	8.36 ± 0.43	6.60 ± 0.17	2.40 ± 0.12				
Cl	31.35 ± 0.75	2.56 ± 0.37	–				
K_2O	7.68 ± 0.16	1.59 ± 0.06	2.02 ± 0.04				
MgO	1.16 ± 0.04	3.94 ± 0.58	1.71 ± 0.03				
TiO_2	1.03 ± 0.14	1.29 ± 0.13	1.02 ± 0.04				
P_2O_5	0.48 ± 0.02	2.89 ± 0.40	0.27 ± 0.04				