



Research article

Stabilization of lead in an alkali-activated municipal solid waste incineration fly ash–Pyrophyllite-based system

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ABSTRACT

This work focuses on the stabilization and speciation of lead (Pb) in a composite solid produced from an alkali-activated municipal solid waste incineration fly ash (MSWIFA)–pyrophyllite-based system. The solid product was synthesized after mixtures of raw materials (dehydrated pyrophyllite, MSWIFA, 14 mol/L aqueous sodium hydroxide, and sodium silicate solution) were cured at 105 °C for 24 h. The product could reduce the leaching of Pb and the Pb concentration in the leachate was 7.0×10^{-3} using the Japanese leaching test and 9.7×10^{-4} mg/L using toxicity characteristics leaching procedure method, which satisfied the respective test criteria and successfully stabilized Pb in this system. The solid product had a compressive strength of 2 MPa and consisted mainly of crystalline phases. Scanning electron microscopy with X-ray analysis and X-ray absorption fine structure suggested that Pb was present along with Al, Si, and O, and that the atomic environment around the Pb was similar to that of PbSiO₃. These results suggest that the alkali-activated MSWIFA–pyrophyllite-based system could be used to stabilize Pb in MSWIFA.

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1. Introduction

Incineration of municipal solid waste (MSW) is widely used to reduce the volume of waste, but results in the production of residues such as bottom ash and fly ash. MSW incineration fly ash (MSWIFA) is one of these residues, and is considered a hazardous waste because it contains toxic substances due to the enrichment of trace amounts of metal in MSW and organic compounds unintentionally generated during the incineration process (Lam et al., 2010; Margallo et al., 2015; Zacco et al., 2014). MSWIFA is composed of fine particles in the submicron range to around 200 μm, although most are under 45 μm (Diaz-Loya et al., 2012; Zacco et al., 2014). MSWIFA commonly contains calcium (Ca), chlorine (Cl), silicon (Si), sodium (Na), potassium (K), aluminum (Al), iron (Fe), and sulfur (S) as major and minor components, as well as toxic metals, such as lead (Pb), zinc (Zn), copper (Cu), chromium (Cr), cadmium (Cd), arsenic (As), and selenium (Se). (Lam et al., 2010; Zacco et al., 2014;

Margallo et al., 2015). MSWIFA is of concern because the properties of the heavy metals in MSWIFA are important for its safe disposal, resource recovery, and beneficial use in construction materials (Zacco et al., 2014; Tang and Steenari, 2016; Funari et al., 2017; Weibel et al., 2017). Pb is an important toxic metal and one of the most prevalent toxic metals in MSWIFA, and its behavior has received much attention (Zacco et al., 2014; Tang and Steenari, 2016; Funari et al., 2017; Weibel et al., 2017; Li et al., 2017).

The solidification and stabilization technique is a popular treatment to reduce the environmental impact of MSWIFA (Lam et al., 2010; Margallo et al., 2015; Zacco et al., 2014). Although Portland cement is widely used for this purpose and is effective in most cases, it is unsuitable for soluble salts and long-term leaching, and has the disadvantages of increasing the volume of waste and CO₂ emissions (Lam et al., 2010; Zacco et al., 2014; Zhang et al., 2017; Provis and Bernal, 2014; Bernal et al., 2016). Currently, several stabilization technologies for MSWIFA are available (Lam et al., 2010; Zacco et al., 2014; Margallo et al., 2015). Chemical stabilization using organic agents, such as chelators, has been considered for the stabilization of heavy metals in MSWIFA, as less material is required and they are more effective than inorganic

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agents. However, organic agents are generally expensive. Heating treatments, such as melting and calcining, can reduce the volume of MSWIFA and encapsulated heavy metals in the residue; however, these techniques require energy for heating and treatment of volatile heavy metals in the flue gas.

Alkali-activated aluminosilicate-based systems produce composite solid products such as geopolymers and zeolite, which have recently been researched as materials for the solidification and stabilization of wastes and heavy metals (Zhang et al., 2017; Provis and Bernal, 2014; Bernal et al., 2016; Johnson and Arshad, 2014). Aluminosilicate minerals, or waste with aluminate and silicate components, are raw materials that react hydrothermally to form solid products under alkali between room temperature to approximately 200 °C, and are considered an environmentally sound technology in terms of reducing CO₂ emissions and energy consumption, because this system does not use cement and does not require the calcination of clinker and limestone to prepare cement. Most heavy metals form hydroxides and are stabilized under alkaline conditions using cement solidification. Conversely, in alkali-activated aluminosilicate-based systems, heavy metals are expected to be involved in the aluminosilicate, resulting in stabilization (Zhang et al., 2017; Provis and Bernal, 2014; Bernal et al., 2016). Most research has used dehydrated kaolinite and coal fly ash as the aluminosilicate in the system and has shown that solid products could solidify MSWIFA or stabilize Pb in some wastes (Zheng et al., 2010; Lancellotti et al., 2009; Galiano et al., 2011; Diaz-Loya et al., 2012; Nikolić et al., 2014; Lee et al., 2016; Li et al., 2015). Other recent studies have used different aluminosilicate minerals as the raw material for alkali activated systems (Bernal et al., 2016; Buchwald et al., 2009; Gharzouni et al., 2016).

Pyrophyllite, Al₂Si₄O₁₀(OH)₂, is an aluminosilicate that consists of layered units comprising an AlO₆ sheet between two SiO₄ sheets (Perez-Rodriguez et al., 2010). Pyrophyllite has various industrial applications, including the production of porcelain, building materials, fire-resistant material, insecticides, textiles, detergents, and cosmetics (Das and Mohanty, 2009; Harvey and Lagaly, 2013; Dondi et al., 2014). Previous studies have examined alkali-activated pyrophyllite-based systems, MacKenzie et al. (2008) reported that the material from alkali-activation of ball-milled pyrophyllite had acceptable hardness and strength. Ikeda (1998) reported that the addition of pyrophyllite to coal fly ash improved the strength of the material after alkali-activation, and that the materials from alkali-activated coal fly ash– and GGBS–pyrophyllite-based systems had sufficient hardness and strength. Mikuni et al. (2005) revealed that pyrophyllite ore containing a large proportion of pyrophyllite could be used as an active filler based on elution experiments using NaOH_{aq}. Kim et al. (2014) reported the synthesis of zeolite Na-X and Na-A using an alkali-activated ground pyrophyllite-based system. Shiota et al. (2017) investigated the formation of pollucite in the material from an alkali-activated MSWIFA–pyrophyllite-based system. However, these studies do not provide sufficient knowledge using them for the solidification of MSWIFA and also have not applied them to the stabilization of toxic metals such as Pb. In addition, there are rich deposits of pyrophyllite, which can support stable generation for various applications (Higashi, 2000; Das and Mohanty, 2009). This suggests that pyrophyllite may be useful as an alternative aluminosilicate source to kaolinite for producing alkali-activated aluminosilicate material.

Therefore, this study examined the effectiveness of an alkali-activated pyrophyllite-based system for the solidification of MSWIFA and stabilization of Pb in it. Our system was a mixture of pyrophyllite, MSWIFA, aqueous sodium hydroxide (NaOH_{aq}), and sodium silicate solution. First, we investigated the properties of the composite solid produced by the system, using a compressive strength test to evaluate hardening of the product, X-ray diffraction

(XRD) to determine its crystal phase, and the Japanese leaching test (JLT-46) and toxicity characteristics leaching procedure (TCLP) method to examine Pb stabilization. The two leaching tests following the JLT-46 and TCLP methods were also conducted for Zn, Cu, Cr, Cd, As, and Se to evaluate the beneficial uses and disposal of products in landfills. We also examined the speciation of Pb in the solid product using scanning electron microscopy with energy dispersive X-ray spectrometry (SEM-EDS) and X-ray absorption fine structure (XAFS). Based on the results, the effective use of this system for MSWIFA was discussed.

2. Materials and methods

2.1. Experimental procedure

MSWIFA (FA for short), collected using a bag filter at a MSWI plant in Japan, was obtained. The exhaust gas from the furnace passed through a boiler and cooling tower, and was then sprayed with slaked lime slurry (0.5–2.0 g/m³) to treat acidic gases before passing through a bag filter. Pyrophyllite (under 350 mesh; Nacalai Tesque, Kyoto, Japan) was dehydrated for 12 h at 900 °C (Perez-Rodriguez et al., 2010). Table 1 shows the elemental compositions of the FA and dehydrated pyrophyllite.

The composite solid product (PyFA) was synthesized in an alkali-activated MSWIFA–pyrophyllite-based system using a method described previously (Zheng et al., 2010; Lancellotti et al., 2009; Galiano et al., 2011; Diaz-Loya et al., 2012; Shiota et al., 2017). First, MSWIFA and the dehydrated pyrophyllite were pre-mixed at 60 rpm for 1 min using an agitator (Tornado; Iuchi Shi-seido, Osaka, Japan). Then, aqueous NaOH solution (special grade; Nacalai Tesque) and a sodium silicate solution (JIS K1408 grade; Nacalai Tesque) were added to the mixture, which was then mixed once at the same rpm in a plastic columnar mold (diameter, 55 mm; height, 71 mm). No Pb compounds were added to the FA. Next, the slurry mixture (with Si/Al and Na/Al molar ratios of 4.4 and 3.4, respectively) was further mixed at 300 rpm for 2 min. After degassing for 24 h, this mixture was cured for 24 h at 105 °C. After

Table 1
Chemical compositions of pyrophyllite and municipal solid waste incineration fly ash (MSWIFA).

Composition		Dehydrated pyrophyllite	FA ^b
SiO ₂	wt%	78.9	11.4
Al ₂ O ₃	wt%	19.3	5.2
TiO ₂	wt%	0.6	0.8
K ₂ O	wt%	0.3	2.7
Fe ₂ O ₃	wt%	0.2	1.0
Na ₂ O	wt%	0.2	4.7
P ₂ O ₅	wt%	0.1	1.1
CaO	wt%	0.1	22.3
MgO	wt%	ND ^a	2.0
CO ₂	wt%	ND ^a	29.0 ^c
SO ₃	wt%	ND ^a	5.9
Cl	wt%	ND ^a	12.0
others	wt%	0.3	0.5
Zn	mg/kg	ND ^a	11000
Pb	mg/kg	ND ^a	820
Cu	mg/kg	ND ^a	695
Cr	mg/kg	ND ^a	310
Cd	mg/kg	ND ^a	106
As	mg/kg	ND ^a	22.5
Se	mg/kg	ND ^a	11.8

^a Not detectable.

^b Municipal solid waste incineration fly ash collected on bag filter with slaked lime slurry sprayed.

^c Organic carbon was not detectable.

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