



Research article

Comparative leaching of six toxic metals from raw and chemically stabilized MSWI fly ash using citric acid



Huawei Wang^{a, b}, Xinxiu Fan^a, Ya-nan Wang^{a, **}, Weihua Li^a, Yingjie Sun^{a, *}, Meili Zhan^c, Guizhi Wu^a

^a College of Environmental and Municipal Engineering, Qingdao University of Technology, Qingdao 266033, PR China

^b Xinjiang Key Laboratory of Environmental Pollution and Bioremediation, Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi 830011, PR China

^c Qingdao MSW Management & Treatment Co. Ltd., Qingdao 266041, PR China

ARTICLE INFO

Article history:

Received 17 August 2017
Received in revised form
22 November 2017
Accepted 26 November 2017

Keywords:

MSWI fly ash
Toxic metals
Leaching behavior
Citric acid
Fraction analysis

ABSTRACT

The leaching behavior of six typical toxic metals (Pb, Zn, Cr, Cd, Cu and Ni) from raw and chemically stabilized (phosphate and chelating agent) municipal solid waste incineration (MSWI) fly ash were investigated using citric acid. Leaching tests indicated that phosphate stabilization can effectively decrease the leaching of Zn, Cd and Cr; whereas chelating agent stabilization shows a strong ability to lower the release of Pb, Cd and Cu, but instead increases the solubility of Zn and Cr at low pH conditions. Sequential extraction results suggested that the leaching of Pb, Zn and Cd in both the stabilized MSWI fly ash samples led to the decrease in Fe/Mn oxide fraction and the increase in exchangeable and carbonate fractions. The leaching of Cr was due to the decrease in exchangeable, carbonate and Fe/Mn oxide fractions in phosphate-stabilized and chelating agent-stabilized MSWI fly ash. The leaching of Cu in both stabilized MSWI fly ash was greatly ascribed to the decrease in Fe/Mn oxide and oxidisable fractions. Moreover, predicted curves by geochemical model indicated that both stabilized MSWI fly ash have the risk of releasing toxic metals under strong acid environment.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

In China, the amount of municipal solid waste (MSW) has increased remarkably, with a total amount of about 19141.9×10^4 tons in 2015, of which approximately 32.26% (6175.5×10^4 tons) was incinerated (www.stats.gov.cn). MSW incineration (MSWI) has many advantages including energy recovery and reduction of waste volume and weight. However, one of the major issues is the generation of considerable amounts of solid residues (i.e., bottom ash and fly ash) (Zhang et al., 2010). MSWI bottom ash with low content of harmful components is mainly used for construction materials (Hjelmar et al., 2007). MSWI fly ash, collected from air pollution control devices, is enriched in various hazardous substances including toxic metals (i.e., Pb, Zn, Cd, Ni, etc.) and dioxins (Eighmy et al., 1995; Okada and Tomikawa, 2012). Therefore, it is considered

as hazardous waste by China Environmental Protection Agency and needs appropriate treatment before further use or being sent to landfills.

The main treatment technologies of MSWI fly ash can be classified into three categories: (1) washing (Chiang and Hu, 2010; Chimenos et al., 2005); (2) chemical stabilization and solidification (Nzihou and Sharrock, 2002; Shiota et al., 2017; Vavva et al., 2017; Zhang et al., 2016a); and (3) thermal treatment (Islam and Park, 2017). The critical drawback of washing treatment using water and acid is the release of soluble salts and heavy metals (Chimenos et al., 2005). Using a melting process to treat MSWI fly ash also has shortcomings such as energy-consumption and equipment corrosion due to the high required melting temperatures (Chiang and Hu, 2010). Traditionally, cement and lime based solidification process has been widely used to stabilize the MSWI fly ash, but it affects the physico-chemical properties of the solidified MSWI fly ash and also greatly increases the volume of the fly ash (Mangialardi et al., 1999). Thus, various chemicals including phosphate (Eighmy et al., 1997; Li et al., 2017), sulfides (Zhao et al., 2002), silicates (Geysen et al., 2004) and chelating agents (Jiang

* Corresponding author.

** Corresponding author.

E-mail addresses: wangyanan1005@yeah.net (Y.-n. Wang), yjsun1971@126.com (Y. Sun).

et al., 2004; Zhang et al., 2016a) have been developed and used for stabilization of MSWI fly ash, and have shown satisfactory results for the immobilization of toxic metals in MSWI fly ash.

One of the most common options for the management of MSWI fly ash is chemical stabilization/solidification followed by disposal in landfills (Vavva et al., 2017). According to the standard for pollution control at the landfill sites of MSW in China, stabilized MSWI fly ash should be separately landfilled in a specific area of MSW landfill. However, at some landfill sites, stabilized MSWI fly ash is landfilled together with MSW due to construction schedules or other problems. Even though less soluble minerals are formed during the chemical stabilization/solidification process, there are serious leaching risks for toxic metals in stabilized MSWI fly ash when MSW and stabilized MSWI fly ash are mixed in landfills. For instance, pH is one of the key factors controlling the leaching behavior of toxic metals. Several studies showed that acidic pH significantly increases the leaching of toxic metals (i.e., Pb, Cu, Zn and Cd) from MSWI fly ash that has been stabilized with orthophosphate, sodium sulfide, lime and chelating agents (Eighmy et al., 1997; Jiang et al., 2004; Komonweeraket et al., 2015; Zhang et al., 2016b). It is worth noting that MSW landfill will generate a large amount of leachate with organic acids (i.e., butyric acid, citric acid and oxalic acid) during the hydrolysis and acidification phases, resulting in low pH environments (Erses and Onay, 2008; Zhao et al., 2000). It is important to understand the effects of organic acids on the leaching of toxic metals from stabilized MSWI fly ash, and the leaching characteristics of toxic metals from different types of chemically stabilized MSWI fly ash. However, few studies have focused on these aspects at present.

In this study, the leaching behaviors of six toxic metals (Pb, Zn, Cr, Cd, Cu and Ni) from raw, phosphate-stabilized and chelating agent-stabilized MSWI fly ash were comparatively investigated using citric acid over a range of pH values from 6.0 to 3.0 to simulate the exposure conditions for stabilized MSWI fly ash in landfills. The mineralogical characterization before and after citric acid leaching was performed by X-ray diffraction (XRD) analysis. In addition, the distribution patterns of toxic metals in raw and stabilized MSWI fly ash during citric acid leaching were investigated by a sequential extraction procedure. The results could help us better understand the potential leaching risk for chemically stabilized MSWI fly ash during mixing with MSW in the landfill, and could provide technological guidance and theoretical support for the proper chemical stabilization of MSWI fly ash.

2. Materials and methods

2.1. Sample collection and property analysis

The fly ash samples used in this study were collected from a MSW incineration power plant in Qingdao city of Shandong province, China. The treatment capacity of this plant is approximately 1500 tons per day. The fly ash was produced from a spray dryer for acid gas neutralization using hydrated lime powder, followed by a fabric filter for the particulate collection. Representative fly ash samples were obtained as follows: 2 kg of fly ash samples were taken eight times a day, for a total 48 kg of samples over 3 days, and then mixed homogeneously. The samples were collected during March 25–28, 2015 and stored at 4 °C until used for experiments (Section 1 and Fig. S1 in supplementary materials).

Two types of chemical stabilizing agents including phosphate and chelating agent were used to stabilize MSWI fly ash. The dosages of phosphate (phosphoric acid, industrial grade, 85%) and chelating agent (sodium diethyldithiocarbamate, industrial grade) for stabilization of toxic metals were optimized in preliminary experiments, and their optimum dosages are mentioned in Section 2

in supplementary materials. After stabilization, 5 kg of the two stabilized samples were dried at room temperature (25 ± 2 °C). The raw fly ash, phosphate-stabilized and chelating agent-stabilized MSWI fly ash samples are denoted as R0, P0, and C0, respectively. The chemical composition of the samples was determined by X-ray fluorescence spectroscopy (XRF) and the results are shown in Table 1. The results indicated that the main components of raw and stabilized MSWI fly ash are CaO, Cl, K₂O, Na₂O, and SO₃. The contents of six typical toxic metals (Pb, Zn, Cr, Cd, Cu and Ni) and other metals (metalloids) were also determined (Table 1).

2.2. Citric acid leaching tests

As the pH in MSW landfills ranges from slightly acidic to neutral pH (Erses and Onay, 2008; Zhao et al., 2000), the effect of solution pH on the leaching of toxic metals was investigated. The pH values ranged from 3.0 to 6.0 to simulate the exposure conditions for stabilized MSWI fly ash in landfills. In addition, a previous report found that the leaching concentrations of toxic metals (i.e., Pb, Zn and Cu) from MSWI fly ash were closely related to the initial pH between 3.0 and 6.0 in 0.1 M citric acid solution (Huang et al., 2011). Therefore, the citric acid solution with initial pH values of 3.0, 4.0, 5.0 and 6.0 were used for the leaching experiment.

The effect of solution pH on the leaching of toxic metals was investigated using batch experiments. Typical liquid to solid ratio of 20:1 was selected in this work and other preliminary tests (Jiang et al., 2009). Citric acid leaching tests were performed in a stoppered 100 ml (total volume) serum bottle by adding 2.000 g fly ash to 40 ml of 0.1 M citric acid leaching solution. The bottles containing samples were shaken on a reciprocating oscillator with a speed of 200 rpm at 25 °C. After shaking for 12 h, all the samples reached the leaching equilibrium as reported by Huang et al. (2011) and Grubb et al. (2000). All experiments were conducted in triplicate. The fly ash samples used for the leaching test had particle sizes of less than 150 μm. The leaching solutions with different pH values of 3.0, 4.0, 5.0 and 6.0 were obtained by adjusting the mixing ratio of citric acid (0.1 M) and sodium citrate (0.1 M) at 0.23:1, 0.69:1, 1.89:1 and 13.28:1, respectively. The labels R1–4 (R1, R2, R3, R4), P1–4 (P1, P2, P3, P4) and C1–4

Table 1
Major chemical composition and toxic metals contents of the MSWI fly ash.

Items	unit	R0	P0	C0
Na ₂ O	%	5.44	4.00	5.26
MgO	%	2.2	2.83	2.29
Al ₂ O ₃	%	1.17	2.18	2.51
SiO ₂	%	3.18	5.17	5.39
P ₂ O ₅	%	0.33	1.89	1.34
SO ₃	%	5.38	5.82	7.16
Cl	%	16.81	14.41	16.84
K ₂ O	%	4.25	3.46	4.19
CaO	%	58.6	57.69	51.95
Fe ₂ O ₃	%	0.84	1.00	1.14
ZnO	%	0.69	0.63	0.67
others	%	1.11	0.93	1.28
Cd	mg/kg	135	128.5	102.3
Pb	mg/kg	1329.2	1170	1252.8
Cu	mg/kg	385.7	361.5	384.2
Cr	mg/kg	150.5	92.7	113.7
Zn	mg/kg	4425	4000	4606.2
Ni	mg/kg	35.0	15.2	24.0
B	mg/kg	9920	8750	9058
Ba	mg/kg	5380	5012	5236
As	mg/kg	165	89.5	78.9
Ti	mg/kg	1870	1906	1859
Mn	mg/kg	375	289	308
Fe	mg/kg	6555	6985	6428
Al	mg/kg	25520	24792	23459

Download English Version:

<https://daneshyari.com/en/article/7478143>

Download Persian Version:

<https://daneshyari.com/article/7478143>

[Daneshyari.com](https://daneshyari.com)