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Research article

Efficiencies of metal separation and recovery in ash-melting of municipal solid waste under non-oxidative atmospheres with different reducing abilities

Takashi Okada*, Hiroki Tomikawa

Laboratory of Solid Waste Disposal Engineering, Graduate School of Engineering, Hokkaido University, Kita 13 Nishi 8, Kita-ku, Sapporo 060-8628, Japan

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ABSTRACT

Ash-melting of municipal solid waste produces molten metal that contains Fe and Cu, and melting furnace fly ash (MFA) that contains Pb and Zn. To recover the metal from the fly ash, Pb and Zn are extracted from the ash by water or enriched in the ash by washing out salts; this separation depends on their leachability. In this study, we investigated the effects of the reducing ability of the atmosphere on the efficiencies of metal separation during melting and metal recovery in water treatment. Different feedstocks (incineration residues) were melted under N_2 or $CO + N_2$ atmospheres. In some of the feedstock materials, volatilization of metallic Cu into MFA was promoted under the atmosphere with greater reducing ability ($CO + N_2$). This increased volatilization inhibited the metal separation in the ash-melting process. Moreover, the higher reducing ability inhibited the formation of water-soluble lead chlorides and decreased the efficiency of metal recovery from the MFA because of the water leaching of the lead compounds. The reducing ability of the atmosphere is difficult to control uniformly in actual ash-melting plants, and we investigated appropriate melting conditions under which the effect of the reducing ability was minimized to promote metal separation and recovery. This minimization was achieved by melting incineration fly ash without additives with Cl gas treatment at 1400 °C.

1. Introduction

Development of melting technique is important for the recovery of metals from waste materials from resource conservation and environmental protection. Wastes containing valuable metals have been subjected to melting processes for metal recovery (Yoshida, 2011), and the incineration residue from municipal solid waste is one of the feedstock materials in such waste-melting systems (Abe et al., 1996; Kinto, 1996; Jung and Osako, 2007; NTS Corporation, 1994). The behaviors of metals in the ash-melting process have been reported (Yoshiie et al., 2002; Jung et al., 2005; Jung and Osako, 2009; Okada and Tomikawa, 2013; Sekito et al., 2014; Li et al., 2015), and these findings are useful to design appropriate waste-melting processes for metal recovery. In the ash-melting process, vapors of Pb and Zn generated from the incineration residue are quenched in the dust collection system in the melting plant, and metal-rich melting furnace fly ash (MFA) is obtained. In the previous studies, high concentration of Pb and Zn (Pb: 9.2 wt% and Zn: 43 wt%) and presence of rare metal in MFA were reported (Okada et al., 2007b; Jung and Osako, 2007). To recover the valuable metals from MFA, chemical leaching methods have been proposed (Kinto, 1996; Izumikawa, 1996; Inoue et al., 1999; Nagib and Inoue, 2000; Fujita et al., 2002; Takaoka et al., 2002; Okada et al., 2007a, b; Alorro et al., 2009). However, before leaching, it is important to produce “high quality MFA,” from which the valuable metals may be more effectively recovered by controlling the conditions during melting.

In MFA production in the ash-melting process, to enhance the metal separation efficiency and obtain metal-rich MFA, it is necessary to leave Fe and Cu in the molten metal and transfer Pb and Zn in MFA. The behaviors of Pb, Zn, Fe, and Cu have been reported to be different depending on the melting conditions (Yoshiie et al., 2002; Jung et al., 2005; Jung and Osako, 2009; Okada and Tomikawa, 2013). The authors performed ash-melting experiments on incineration residues with different chemical compositions at different melting temperatures, changing the atmosphere from oxidative to non-oxidative (Okada and Tomikawa, 2013).

* Corresponding author. Present address: Headquarters for Innovative Society-Academic Cooperation, University of Fukui, Bunkyo 3-9-1, Fukui 910-8507, Japan.
E-mail address: t-okada@u-fukui.ac.jp (T. Okada).

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Table 1
Chemical compositions of the IFA samples and the BA sample obtained. (Note. Bold italics means that concentrations of Na and Ca are high).

Element	Content (wt%)			
	IFA_{Ca}	IFA_{Na}	IFA_{N}	BA
Zn	0.87	0.77	2.4	0.30
Pb	0.18	0.24	0.48	0.055
Fe	0.38	0.81	0.87	2.0
Cu	0.06	0.05	0.13	0.46
Na	5.4	***20***	7.3	2.3
K	8.2	9.0	8.3	1.3
Cl	25	26	17	1.2
Ca	***22***	7.2	13	16
Si	6.3	4.6	8.9	21
H	0.96	0.25	0.41	0.23
C	1.4	4.0	6.1	1.7
Al	1.5	2.0	3.0	5.6
S	1.7	2.5	2.7	0.40
Mg	0.76	0.79	1.3	1.2
Ti	0.28	0.72	0.88	1.2
P	0.45	0.41	0.87	1.9
Cr	0.0045	0.041	0.019	0.011
Cd	0.0087	0.0041	0.039	0.00045
Sb	0.045	0.029	0.10	0.0092
Mn	0.0050	0.023	0.058	0.11

Based on the results, melting conditions necessary to achieve a high metal separation efficiency were proposed.

Another key aspect of MFA production concerns the leaching characteristics of Pb and Zn from the resulting MFA, which determine the metal recovery efficiency. In MFA, Na, K, and Cl are also concentrated, and the heavy metals in MFA were contaminated by the salts (Jung et al., 2005); such salts should be washed out with water to concentrate Pb and Zn (Okada et al., 2007b). In this water treatment, compounds of Pb and Zn with water-soluble forms are extracted from MFA in water (water-leaching method). The extracted Pb and Zn are precipitated as their sulfides or hydroxides, or as metal. The extracted Na, K, and Cl remain in the liquid phase. On the other hand, compounds of Pb and Zn with water-insoluble forms are left behind in the MFA and enriched (water washing method). To effectively recover Pb and Zn from the MFA by water treatment, the melting conditions should be controlled to render the chemical forms of Pb and Zn as either all water-soluble or all water-insoluble. To investigate such conditions, we evaluated the leaching characteristics of Pb in MFA generated by melting different incineration residues at various temperatures, in both oxidative and non-oxidative atmospheres (Okada and Tomikawa, 2012).

Based on our aforementioned studies (Okada and Tomikawa, 2012, 2013), a non-oxidative atmosphere facilitates metal separation in the melting process and metal recovery in water treatment. However, the reducing ability of a non-oxidative atmosphere would vary between actual ash-melting plants. In a coke bed-type furnace, the feedstock is melted with coke, and CO is generated by combustion of the latter. In arc-, plasma-, and electric-resistance-type furnaces, graphite electrodes are used for heating and CO is generated by electrode combustion. The partial pressures of CO in the melting furnaces therefore vary from plant to plant. Therefore, it is important to evaluate the effects of the reducing ability of the atmosphere on the efficiencies of metal separation and recovery, as mentioned above. To the best of our knowledge, these effects have not been investigated in previous studies.

In this study, we investigated the effects of the reducing ability of the melting atmosphere on the two types of efficiencies: 1) that of the separation of Pb and Zn from Cu and Fe in the melting process, and 2) that of the recovery of Pb and Zn from the generated MFA in water treatment. The incineration residues were melted in a laboratory-scale reactor, under atmospheres in the reactor of N_2 and $CO + N_2$ (higher reducing ability). The metal separation efficiency was evaluated based on the amount of metal transferred to the generated MFA, and that of the metal recovery from water treatment was determined based on the leaching characteristics of Pb and Zn in the MFA.

2. Materials and methods

We previously used one incineration bottom ash (BA) sample and three incineration fly ash (IFA) samples with different chemical compositions in melting experiments (Okada and Tomikawa, 2012, 2013). Table 1 shows the chemical compositions of the BA and IFA samples, which were also used in the present study. The analytical methods of the chemical compositions are explained below. By the differential thermal conductivity technique, concentrations of C and H in the ashes were determined. The concentrations of Cl and S were determined as follows. The ashes were burned in a flask that contains oxygen and a mixed solution of $NaHCO_3$, Na_2CO_3, and H_2O_2. The obtained solutions were analyzed by ion chromatography and the concentrations of Cl^- and SO_4^{2-} in the solutions were determined. To determine the concentrations of P in the

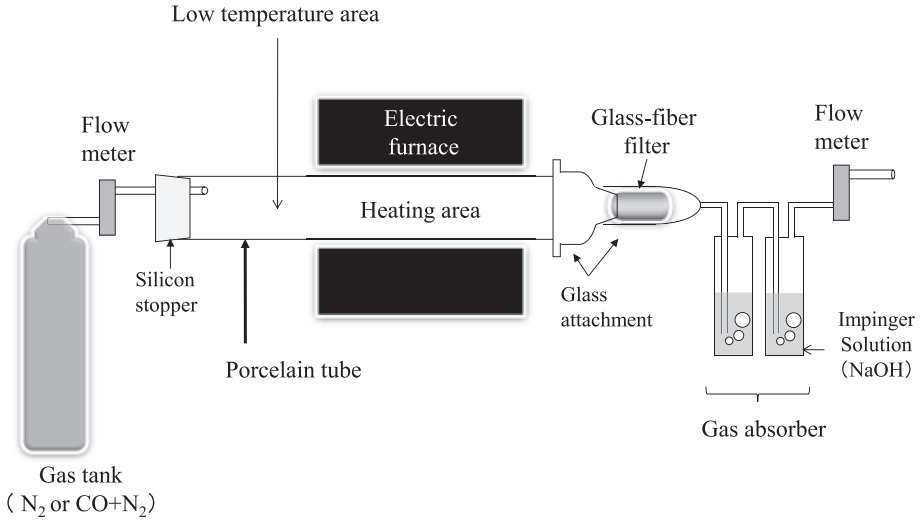

Fig. 1. Schematic diagram of the lab-scale reactor.