



Leaching characteristics of lead from melting furnace fly ash generated by melting of incineration fly ash

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ABSTRACT

This study investigated the effect of the chemical composition of incineration fly ash on the leaching characteristics of Pb from melting furnace fly ash generated by melting incineration fly ash. Melting furnace fly ash from both a real-scale melting process and lab-scale melting experiments was analyzed. In addition, the theoretical behavior of Cl that affects the leaching characteristics of Pb was simulated by a thermodynamic equilibrium calculation. Proportions of water-soluble Pb in the melting furnace fly ash were correlated with equivalent ratios of total Pb in the ash and Cl transferred to gas. The amount of Cl in the gas increased with an increase in the molar ratio of Cl to Na and K in the incineration fly ash. The thermodynamic calculation predicted that HCl generation is promoted by the increase in the molar ratio, and X-ray photoelectron spectroscopy indicated a possible presence of $PbCl_2$ in the melting furnace fly ash. These results implied that the formation of water-soluble $PbCl_2$ with HCl was affected by the relationships among the amounts of Na, K, and Cl in the incineration fly ash. This is highly significant in determining the leaching characteristics of Pb from the melting furnace fly ash.

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1. Introduction

The melting technique is one available treatment option in solid waste management systems. In waste melting processes, some valuable metals in the waste are volatilized and metal-rich materials, known as melting furnace fly ash (MFA), are produced. Contents of Pb and Zn in MFA have been reported to be up to 9.2 wt% and 43 wt%, respectively (Okada et al., 2007b), and it was reported that rare metals were also found in MFA (Jung and Osako, 2007 and 2009a). Because of the ability of MFA to concentrate metals, the melting technique can be used for metal recovery from multicomponent end-of-life products (waste) in Japan, where conserving resources is of particular interest with respect to sustainable development and environmental protection.

Among the multicomponent waste products, incineration fly ash (IFA) contains higher amounts of heavy metals than bottom ash (BA), and would be preferred for the recovery of metals, especially Pb and Zn. However, as metal concentrations are still relatively low in IFA for the purposes of metal recovery (Okada et al., 2007b), it is

necessary to extract the heavy metals and concentrate them. The melting technique can therefore be applied to IFA to achieve metal concentration. Unfortunately, large quantities of salts (Na, K, and Cl) in IFA are also volatilized to MFA and they dilute the content of the heavy metals (Jung et al., 2005). To separate the heavy metals from the salts, water treatment or chemical leaching can be used (Kinto, 1996; Izumikawa, 1996; Inoue et al., 1999; Nagib and Inoue, 2000; Fujita et al., 2002; Takaoka et al., 2002; Okada et al., 2007a, 2007b; Alorro et al., 2009). In the chemical leaching, the heavy metals are extracted from MFA into chemical solvents, and the extracted metals are recovered by precipitation as a sulfide, a hydroxide or a metal. The water treatment is both simpler and more cost-effective. Water leaching and water washing are the water treatments generally used. In water leaching, the heavy metals and salts are extracted from MFA by water, and then the extracted heavy metals are recovered by the precipitation, leaving behind the extracted salts in the liquid phase. When heavy metals are present as water-soluble compounds in MFA, the water leaching technique is effective for the treatment of MFA. In the water washing technique, the salts are removed with water to leave behind the heavy metals in MFA, and these heavy metals can then be enriched. Thus, when heavy metals exist as water-insoluble compounds, the water washing technique should be used. Therefore, the leaching characteristics of heavy metals in MFA are important in selecting the most effective water treatment method. Jung and Osako (2009b)

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investigated the leaching characteristics of metals present in MFA generated from ash-melting plants where the proportions of IFA in the feedstock were different. The proportion of IFA is important in determining the leaching characteristics because large amounts of Cl are present in IFA and they promote the formation of water-soluble metal chlorides in the melting of IFA. However, IFA has different chemical compositions depending on the exhaust gas treatment system used in the incinerator, a consideration which was not discussed by Jung and Osako in relation to its effects on leaching characteristics. This issue has not been discussed in previous studies of the ash-melting process.

Our objective, therefore, was to investigate the effects of the chemical composition of IFA on the leaching characteristics of Pb from MFA generated by melting IFA. MFA generated from both a real-scale melting process and a lab-scale melting experiment was analyzed to evaluate the leaching characteristics of Pb. In addition, the experimental results were considered theoretically based on a thermodynamic equilibrium calculation.

2. Materials and methods

2.1. Investigation of MFA generated from a real-scale melting process

The leaching characteristics of Pb can be affected by the chemical forms of Pb in MFA, and these chemical forms depend on chemical compositions of feedstock. Therefore, the leachability and speciation of Pb in MFA from a real-scale melting process were evaluated in order to investigate the effects of the chemical composition of IFA. Table 1 provides general information regarding the ash-melting plant, from where we collected the MFA sample. The plant is equipped with an electric resistance furnace in which an electrode is used for heating, and IFA is melted under a non-oxidative atmosphere. Table 2 shows the chemical composition of the MFA sample collected. The main components of the sample were Na, K, and Cl, with lower levels of Pb and Zn. In most ash-melting plants, $\text{Ca}(\text{OH})_2$ is injected into the exhaust gases from the melting furnaces to neutralize HCl in the gases, and the injected Ca is mixed with MFA. In the melting plant we selected, however, the $\text{Ca}(\text{OH})_2$ injection was performed after collecting MFA, and the MFA sample therefore did not contain Ca originating from the injection.

A water leaching test was performed on the MFA sample to evaluate the leaching characteristics of Pb from the sample. In the test, 2.5 g of MFA was charged into a glass flask containing 100 mL of distilled water. The flask was sealed, and then shaken horizontally at room temperature for 1 h, with a shaking frequency of 200 min^{-1} . After shaking, the solution was separated from the solid by vacuum filtration using a $0.1\text{-}\mu\text{m}$ membrane filter, and the Pb concentration in the solution obtained was measured by atomic absorption spectrometry (AAS) on a Hitachi Z-8200 system (Hitachi, Tokyo, Japan).

To evaluate the speciation of Pb in the MFA sample, the chemical state of Pb was analyzed by X-ray photoelectron spectroscopy (XPS) using an AXIS-His (Shimadzu/Kratos) X-ray photoelectron spectrometer. Inano et al. (2008) applied this technique to MFA. In our analysis, a monochromated Al K α source ($h\nu = 1486.6 \text{ eV}$) was used in conjunction with a 127-mm radius concentric hemispherical

Table 2

Chemical composition of the MFA sample from the practical melting plant.

| Element | Content (wt%) |
|---------|---------------|
| Zn | 0.27 |
| Pb | 0.44 |
| Fe | 0.013 |
| Cu | 0.15 |
| Na | 13 |
| K | 14 |
| Cl | 37 |
| Ca | 0.24 |
| Al | 0.015 |
| S | 1.6 |
| H | 0.11 |
| C | 0.24 |
| Cr | 0.053 |
| Cd | 0.015 |

analyzer, and low energy electron flux was irradiated by a charge neutralizer to avoid a buildup of charge. The absolute binding energy of the photoelectron spectra was determined by referencing to the C 1 s transition at 284.6 eV, which results from background hydrocarbons in the ultrahigh vacuum environment.

2.2. Lab-scale experiment of ash melting

In real-scale plants, it is difficult to compare melting processes of IFA with different chemical compositions under the same melting conditions (melting temperature, atmosphere, and residence time) because the conditions and feedstock differ from plant to plant (Jung et al., 2005). Therefore, IFA samples with different chemical compositions were also melted in a lab-scale reactor, allowing us to control the melting conditions.

2.2.1. Materials

Three IFA samples were obtained from stoker-type incineration plants (Plant A, B, and C) of municipal solid waste (MSW) with different exhaust gas treatment systems. In Plant A, $\text{Ca}(\text{OH})_2$ is injected into an exhaust gas from the incinerator to neutralize HCl, and IFA is collected after the injection. Therefore, the collected IFA (IFA_{Ca}) contains large quantities of Ca originating from the $\text{Ca}(\text{OH})_2$. In Plant B, NaHCO_3 is used for the neutralization of HCl, and large amounts of Na are present in the collected IFA (IFA_{Na}). The third IFA sample (IFA_{N}) was obtained from Plant C where the IFA is collected before the $\text{Ca}(\text{OH})_2$ injection, and IFA_{N} does not contain any additives. In addition, one BA sample was obtained from an MSW incineration plant. Although corresponding IFA and BA from a same incinerator are melted together in the practical ash-melting process, one BA sample from the separate combustion unit was melted with each of the IFA samples in this study (see Section 2.2.3) to avoid differences in BA composition. Table 3 shows the chemical compositions of the samples obtained. The concentrations of Ca and Na differed from IFA to IFA.

2.2.2. Apparatus

Fig. 1 is a schematic of the lab-scale reactor consisting of a gas tank, flow meters, a porcelain tube, an electric tubular furnace, glass attachments, and a gas absorber. The gas tank contained air or N_2 , and provided a continuous gas flow through the porcelain tube to control the atmosphere in the tube. The flow meter was connected between the tank and the tube to check the gas flow rate. The porcelain tube was placed inside the electric tubular furnace to heat the sample during the melting experiment. A $1\text{-}\mu\text{m}$ glass-fiber filter was inserted in the glass attachments to collect the MFA generated. The gas absorber consisted of two glass containers, both of which

Table 1

Characteristics of the melting plant studied.

| Furnace type | Capacity ($t \text{ day}^{-1}$) $\times n$ | Atmospheric condition | Feedstock |
|---------------------|---|--------------------------|-----------|
| Electric resistance | 4.8×1 | Non-oxidative | IFA |

Table 3
Chemical compositions of the IFA samples and the BA sample.

| 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 |
| 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 |
| P | 0.45 | 0.41 | 0.87 | 1.9 |
| H | 0.96 | 0.25 | 0.41 | 0.23 |
| C | 1.4 | 4.0 | 6.1 | 1.7 |
| 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 |
| Ti | 0.28 | 0.72 | 0.88 | 1.2 |
| Mn | 0.0050 | 0.023 | 0.058 | 0.11 |

Note. Italics means that concentrations of Na and Ca are high.

contained 50 mL of impinger solution (1.0 wt% NaOH), to absorb the generated gas passing through the filter. The flow meter was connected to the exit pipe of the absorber to check the gas flow rate.

2.2.3. Experimental conditions and procedure

Using the IFA samples and the BA sample, three types of test samples were prepared. Each of the IFA samples was mixed with the BA sample because IFA is generally melted with BA in practice. Mass ratios of IFA to total feedstock have previously been reported to be 0.2–0.5 (Okada et al., 2007a), and a ratio of 0.5 was therefore chosen to clearly determine the effect of IFA. The mixture samples were labeled IFA_{Ca} + BA, IFA_{Na} + BA, and IFA_N + BA. In the experiments, gas purging conditions and melting temperatures were varied to investigate the effects of different combinations with the chemical composition of IFA on the leaching characteristics of Pb from the MFA generated. The atmosphere in a real ash-melting furnace ranges from non-oxidative to oxidative depending on the type of melting furnace (Jung and Osako, 2007), and the gases used in our experiments were therefore N₂ (non-oxidative) and air

(oxidative). Melting temperatures in real ash-melting furnaces were 1300–1750 °C (Jung et al., 2005), and the temperature limit of the electric tubular furnace used in this work was 1500 °C. Therefore, the melting temperatures examined in this study were 1300 °C, 1400 °C, and 1500 °C.

In the melting experiment, 5 g of the test sample was loaded in a porcelain crucible, which was then placed in a low temperature area in the porcelain tube (see Fig. 1). The tube was heated to a given temperature by the furnace, and then air or N₂ was supplied into the tube at a flow rate of 1.5 L min^{−1}. After a holding time of 30 min, the crucible was moved to the heating area in the tube (see Fig. 1) in order to heat the sample under gas purging conditions. During heating the sample, air or N₂ was kept supplied into the tube at a flow rate of 1.5 L min^{−1}. The MFA generated was collected by the filter and the generated gas was bubbled through the impinger solution in the gas absorber. After 1 h, the tube naturally cooled down to room temperature, and the MFA collected by the filter and impinger solution was obtained.

The MFA collected with the filter (1.7–2.6 g) was charged into a glass flask containing 100 mL of distilled water, and the leaching test was performed in the manner explained in Section 2.1. The residue after the leaching test was put into a glass flask containing 120 mL of aqua regia, and digested by heating at 110 °C for 24 h. After digestion, the solid–liquid separation and the analysis of the Pb concentration in the filtrate were performed, as explained in Section 2.1. In addition, the Cl concentration in the impinger solution, the discussion of which is significant for the leaching characteristics of Pb (see Section 3.2.1), was measured using the Hg(SCN)₂ method.

2.3. Equilibrium calculation method

Thermodynamic equilibrium calculations were performed using the FactSage 5.2 program (GTT Technologies) (Bale et al., 2002). This program provides equilibrium calculation data for a system of maximum 693 components, which is useful for predicting the dominant species of metals in a multicomponent and multiphase system constituted by BA and IFA. As described in Section 3.2.1, Cl behavior in the melting process is relevant to the leaching characteristics of Pb from MFA, and the dominant species of Cl was predicted by the calculation. Jung and Osako (2007) also used this program for a similar prediction in the ash-melting processes.

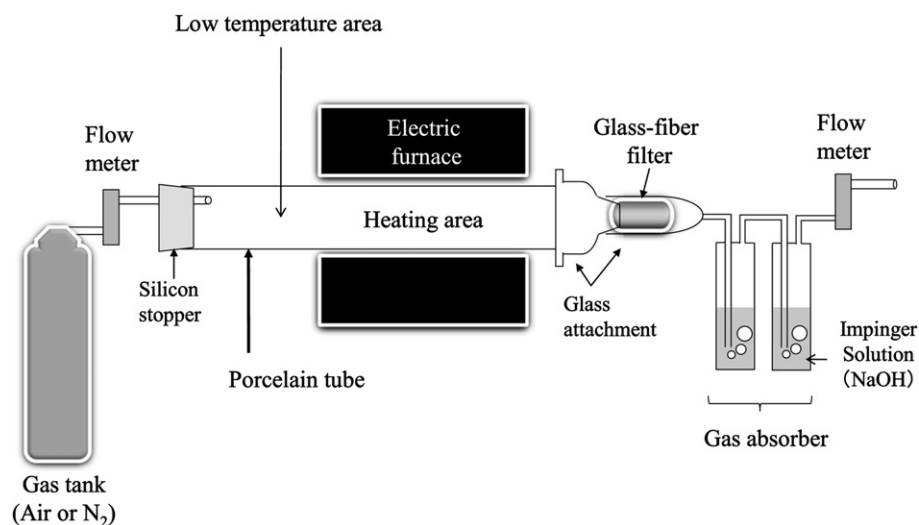


Fig. 1. Lab-scale reactor. The gas tank, the flow meters, the porcelain tube, the electric tubular furnace, the glass attachments, and the gas absorber used in the melting experiment are illustrated.

Table 4

Chemical compositions of the initial values for thermodynamic calculation (unit: mol kg⁻¹).

| | IFACa + BA | IFANa + BA | IFAN + BA | BA |
|----|------------|------------|-----------|----------|
| O | 20 | 19 | 21 | 26 |
| Zn | 0.089 | 0.081 | 0.21 | 0.045 |
| Pb | 0.0056 | 0.0072 | 0.013 | 0.0026 |
| Fe | 0.21 | 0.25 | 0.26 | 0.36 |
| Cu | 0.041 | 0.040 | 0.047 | 0.072 |
| Na | 1.7 | 4.7 | 2.1 | 1.0 |
| K | 1.2 | 1.3 | 1.2 | 0.33 |
| Cl | 3.7 | 3.9 | 2.5 | 0.34 |
| Ca | 4.7 | 2.9 | 3.6 | 4.0 |
| Si | 4.8 | 4.5 | 5.3 | 7.4 |
| H | 5.9 | 2.4 | 3.2 | 2.3 |
| C | 1.3 | 2.4 | 3.2 | 1.4 |
| Al | 1.3 | 1.4 | 1.6 | 2.1 |
| S | 0.32 | 0.45 | 0.49 | 0.13 |
| Mg | 0.40 | 0.41 | 0.51 | 0.49 |
| Ti | 0.15 | 0.20 | 0.21 | 0.25 |
| P | 0.38 | 0.38 | 0.45 | 0.62 |
| Cr | 0.0014 | 0.0050 | 0.0029 | 0.0020 |
| Cd | 0.00040 | 0.00020 | 0.0017 | 0.000040 |
| Sb | 0.0022 | 0.0016 | 0.0046 | 0.00075 |
| Mn | 0.010 | 0.012 | 0.015 | 0.019 |

Table 4 shows the input data on a mol kg⁻¹ basis. In the calculation, 21 elements that occupy 97% in the feedstock were selected to keep the number of product components below its limitation (<693) in the FactSage program. The O-contents used were calculated using the metal content, assuming that the metals were present in the form of oxides. Although this is just an estimation, the O-contents were similar to those used by Jung and Osako (2007). The examined melting temperatures were set at 1300 °C, 1400 °C, and 1500 °C, and the atmospheres were changed from non-oxidative to oxidative. In the oxidative atmosphere, the calculation was performed with 17 mol kg-ash⁻¹ O₂, which is the same amount purged into the reactor in the experiment. In the calculation, a slag solution was simulated using a slag model in the FactSage program. The slag model should include metal chlorides to simulate the theoretical behaviors of Cl, and therefore the slag solution employed was a mixture of oxides (MgO, FeO, SiO₂, CaO, Al₂O₃, Fe₂O₃, PbO, ZnO, and Cu₂O), sulfides (MgS, CaS, FeS, ZnS, PbS, and Cu₂S), and chlorides (CaCl₂, MgCl₂, FeCl₂, PbCl₂, ZnCl₂, and CuCl).

3. Results and discussion

3.1. Leachability and speciation of Pb present in MFA from real-scale melting process

Before comparing the effects of the chemical compositions of IFA, the leachability and speciation of Pb in MFA from the real-scale melting process were confirmed. Table 5 shows the results of the leaching test performed on the MFA sample generated from the practical melting plant. The Pb extraction described in Table 5 is defined as follows.

$$\text{Pb extraction (\%)} = M_L^{\text{Pb}} / M_M^{\text{Pb}} \times 100,$$

where M_L^{Pb} is the amount of Pb extracted into distilled water in the

Table 5

Results of the leaching test performed on the MFA sample from the practical melting plant.

| Pb extraction (%) | Solution pH | Pb/water ratio ^a (mg L ⁻¹) |
|-------------------|-------------|---|
| 85 | 5.3 | 110 |

^a The ratio of the Pb amount in the MFA sample to water volume.

leaching test and M_M^{Pb} is the amount of Pb present in the MFA sample.

It was found that 85% of Pb present in the MFA sample was extracted into distilled water. This water-soluble Pb could be recovered by the water leaching method, but the remaining Pb would stay unrecovered in the MFA. In the leaching test, the ratio of the amount of Pb in the MFA sample to water volume was 110 mg L⁻¹, and the pH of the solution after extraction was 5.3. At this pH, the theoretical solubility of Pb was calculated using the method proposed by Sakanakura (1999), assuming that the concentration of inorganic carbon in the solution is 0.22 mg L⁻¹ (saturated solubility). The solubility of Pb calculated was 600 mg L⁻¹ (>110 mg L⁻¹). One possible explanation for the difference between the water-soluble Pb and the remaining Pb is the difference in the chemical forms of Pb.

Fig. 2 shows the analytical results for Pb speciation by the XPS analysis performed on the MFA sample. From the different Pb photoelectron peaks observed in the analysis, we selected a 4f spectrum with a high signal intensity and a distinct chemical shift. As a reference, standard Pb reagents (Pb, PbCl₂, PbSO₄, PbO, PbO₂, PbO₃, PbS, and PbSiO₃) were also analyzed, and the binding energy at the peak position for each reagent is described together with the result of the MFA sample in Fig. 2. The peak position for MFA was close to those for PbCl₂ and PbCO₃. The solubility of PbCl₂ in water is 0.98 g 100 mL⁻¹ at 20 °C (Lide, 2003), while that of PbCO₃ is 0.00011 g 100 mL⁻¹ at 20 °C (Editorial committee of dictionary of chemistry, 1963). In Section 3.2, the leaching characteristics of Pb from MFA will be discussed assuming that the water-soluble Pb in MFA is PbCl₂.

3.2. Effects of chemical compositions of IFA on leaching characteristics of Pb from MFA

3.2.1. Results of lab-scale experiment

Before discussing the leaching characteristics, the accuracy of the composition data of the ash samples (IFACa, IFANa, IFAN, and BA) is checked. The content of Pb in each sample was determined with the digestion method explained in Section 2.2.3, and the analysis

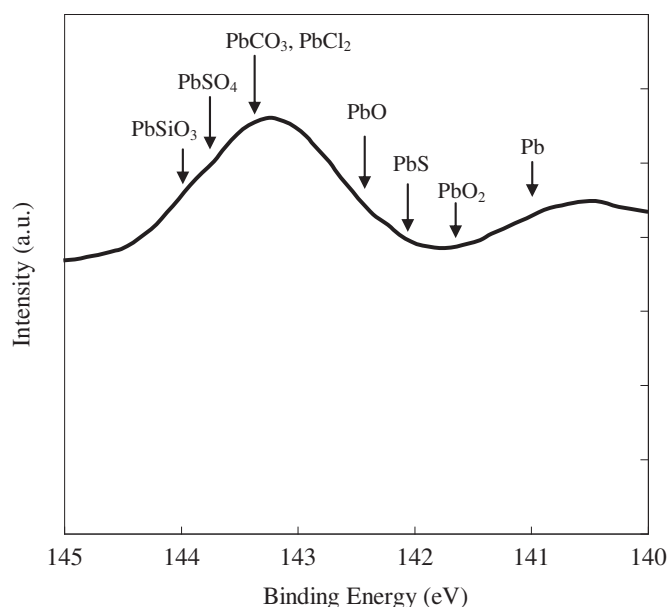


Fig. 2. Analysis of the X-ray photoelectron spectroscopy. Pb4f 5/2 photoelectron spectra are shown for the MFA sample, and the analytical results of the standard Pb reagents (PbSiO₃, PbSO₄, PbCO₃, PbCl₂, PbO, PbS, PbO₂, and Pb) are added.

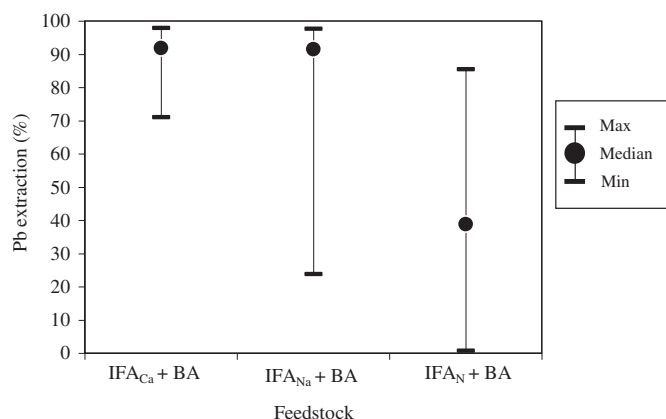


Fig. 3. Leaching characteristics of Pb from MFA. The median values and ranges of the Pb extractions are shown for each feedstock.

was repeated twice. The coefficients of variations were 14% for IFA_{Ca}, 0.77% for IFA_{Na}, 0.0084% for IFA_N, and 39% for BA.

Fig. 3 shows the effects of the IFA types on the leaching characteristics of Pb from the MFA generated by the lab-scale melting experiments. The melting experiments were performed under different melting temperatures and atmospheres, and the Pb extractions varied widely in each of the IFA types. In Fig. 3, median values and ranges of the Pb extractions in all the experiments are presented. The median values and ranges differed depending on the IFA type, and tended to follow the order IFA_{Ca} > IFA_{Na} > IFA_N.

Next, the effects of different combinations of IFA types with melting temperature or atmosphere are discussed. Fig. 4 shows the effects of the melting temperature on the Pb extractions. In the case where the IFA_{Ca} sample was melted, the Pb extractions increased with decreasing temperature. On the other hand, this trend was reversed when the IFA_{Na} sample or IFA_N sample was melted. Fig. 5 shows the effects of the atmosphere on the Pb extractions. As the atmosphere became oxidative, the Pb extractions increased. The increase in the Pb extractions by the oxidative atmosphere was highest for IFA_N among the IFA types.

The difference in Pb extractions is considered below, in which it is assumed that these differences were caused by the difference in the amounts of PbCl₂ in MFA, as mentioned in Section 3.1. In the melting process, the formation of PbCl₂ can be affected by the amount of Cl in the gas phase. If the water-soluble Pb originates from PbCl₂, there is a relationship between the Pb extractions and the Cl concentrations. Therefore, the amount of Cl transferred to the gas after collecting MFA was determined by analyzing the impinger solution. These Cl concentrations are used in the following analysis, and equivalent ratios of the amount of Cl to the amount of Pb in MFA (Cl-gas/Pb ratio) is defined as follows.

$$\text{Cl-gas/Pb ratio} = M_g^{\text{Cl}} / M_M^{\text{Pb}},$$

where M_g^{Cl} is the amount of Cl in the gas (impinger solution) and M_M^{Pb} is the amount of Pb in MFA.

Fig. 6 shows the relationship between the Cl-gas/Pb ratios and the Pb extractions. When the Cl-gas/Pb ratio ranged from 0 to 30, the Pb extractions increased with increasing Cl-gas/Pb ratio. As the Cl-gas/Pb ratio increased further, over 90% of the Pb present in MFA

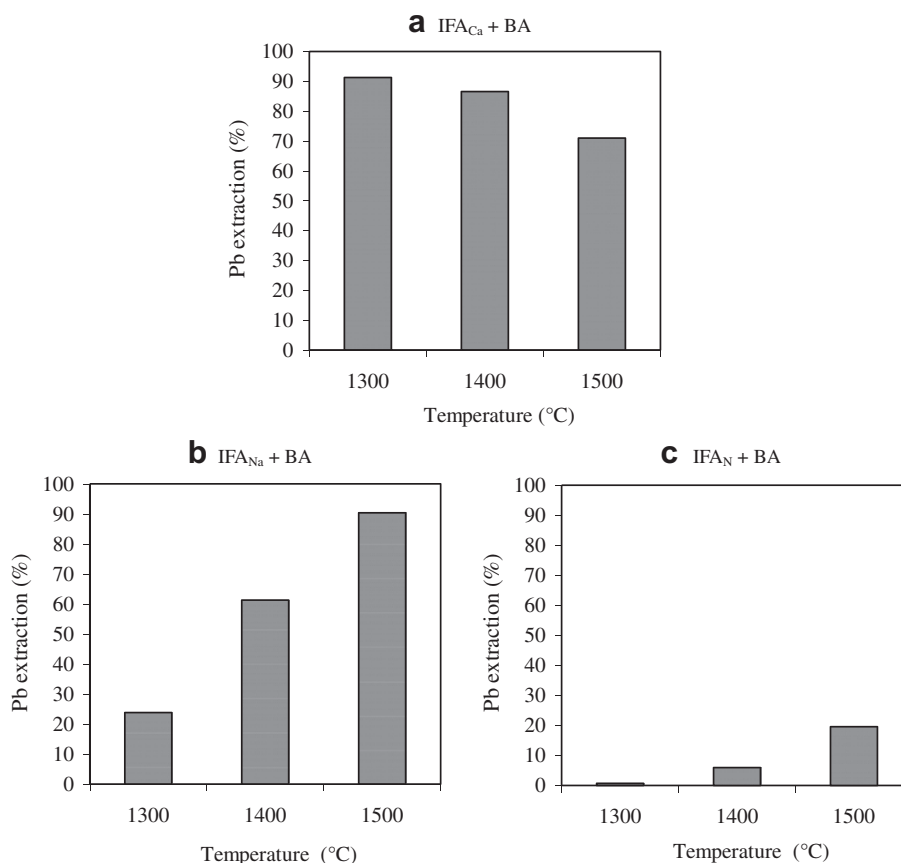


Fig. 4. Effect of the temperature on the leaching characteristics of Pb from MFA. Relationships between the Pb extractions and the temperatures are shown for each feedstock.

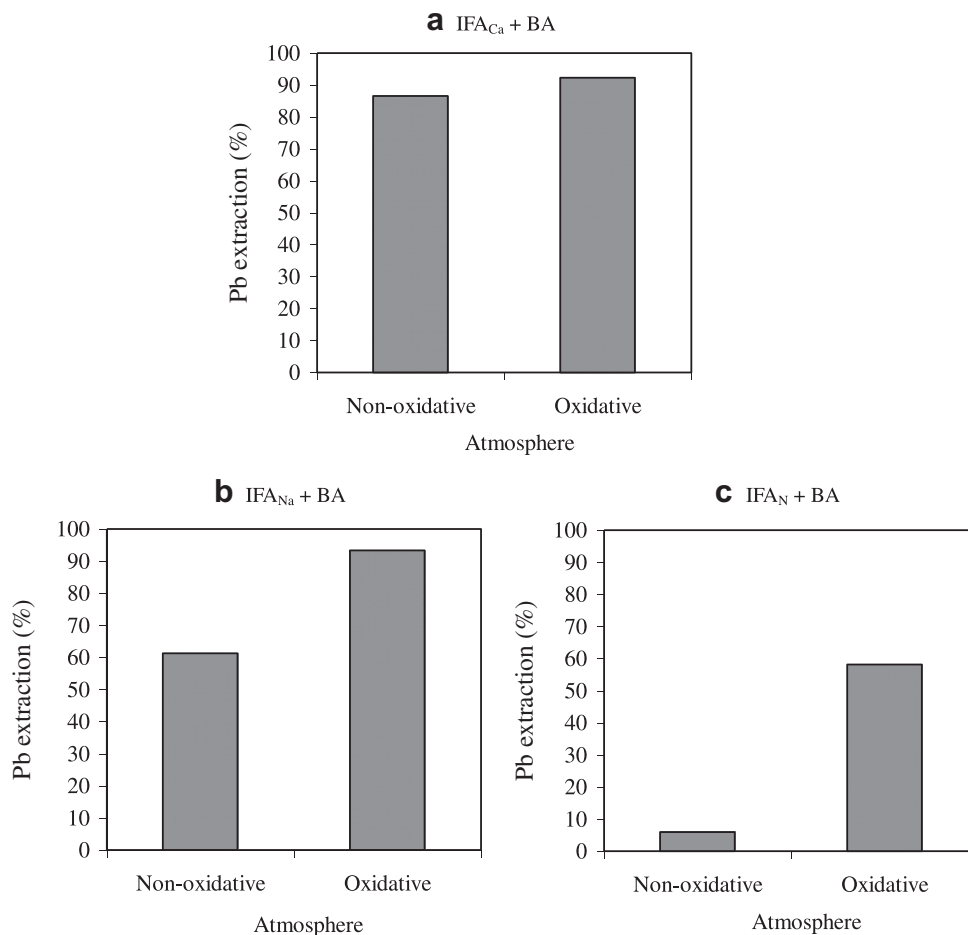


Fig. 5. Effect of the atmosphere on the leaching characteristics of Pb from MFA. Relationships between the Pb extractions and the atmospheres are shown for each feedstock.

was extracted into distilled water. The chemical reaction of Pb and Cl cannot be confirmed by the correlation, but it can be concluded that the amount of Cl in the gas is an influential factor in determining the leaching characteristics of Pb from MFA. The Cl amount in the gas can be affected by the chemical composition of IFA, as explained below. Fig. 7 shows Ellingham diagrams for the chlorination reaction of the oxides of the main components (Na, K, Ca, and Si) in the feedstock, described using thermodynamic data from previous investigations (Knacke et al., 1991). After the condensation of the metal vapors in the MFA collection, HCl is a possible gaseous Cl species, and the Gibbs free energies of the chlorination reaction of H₂O is represented in Fig. 7. Although the Gibbs free energy for each reaction is not fully described in all temperature ranges, it can be considered that the affinities of Na and K to Cl are higher than those of the other elements, as indicated by the lower Gibbs free energies of the chloride formations of Na and K than those for chlorides of the other elements. Furthermore, in the ash-melting process, Ca was thermodynamically stable as CaO in the slag phase, as mentioned in Section 3.2.2. Therefore, if the amount of Cl is greater than those of Na and K in the ash-melting system, it is more likely that Cl combines with H. The relationship among the amounts of Na, K, and Cl in the feedstock differed among IFA types, as can be seen in Table 3. Fig. 8 shows the effects of the difference in the relationship on the amount of Cl transferred to the gas. The horizontal axis in Fig. 8 indicates the molar ratios of Cl to Na and K in the feedstock (Cl/(Na + K) ratio). The Cl/(Na + K) ratio in the BA sample was much lower than those in the IFA samples, and the result of the experiment in which only the BA sample was melted is

represented in Fig. 8 to clearly show the effects of the Cl/(Na + K) ratio. In both the oxidative and non-oxidative atmosphere, the amounts of Cl transferred to the gases were positively correlated with the Cl/(Na + K) ratios, and the gradient was higher in the oxidative atmosphere.

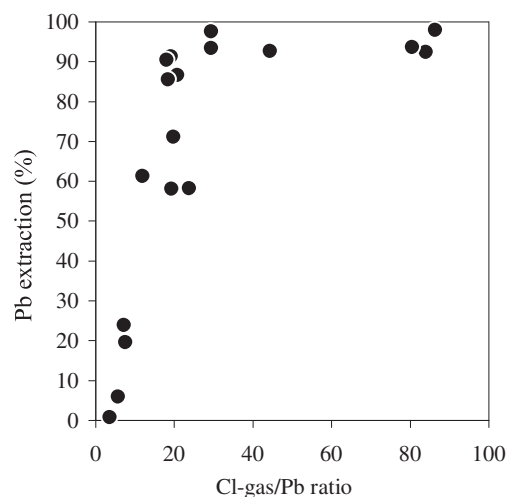


Fig. 6. Relationship between the Pb extractions and Cl-gas/Pb ratios. Vertical axis shows the Pb extractions, and horizontal axis does the equivalent ratios of Cl in the gas to Pb in MFA generated by the melting experiments.

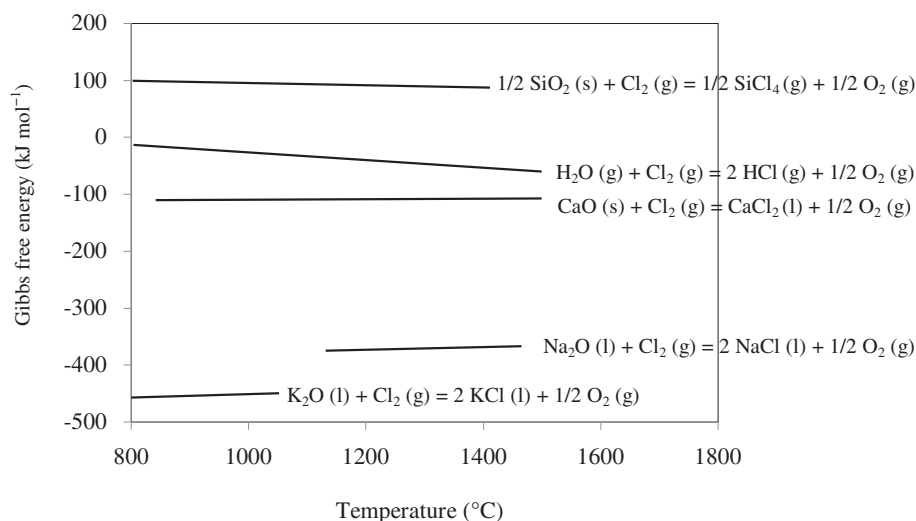


Fig. 7. Ellingham diagrams. Gibbs free energies at 800–1500 °C are shown for the chlorination-reactions of SiO₂, H₂O, CaO, Na₂O, and K₂O.

3.2.2. Thermodynamic behaviors of Cl

The relationship between the Cl-gas generations and the Cl/(Na + K) ratios is theoretically simulated by the thermodynamic equilibrium calculation to support the discussion of the Cl affinity based on the Ellingham diagram (Fig. 7) in Section 3.2.1. Fig. 9(a) summarizes phase distributions of Cl simulated by the calculation. Most of Cl in the gas phase was predicted to be present as NaCl, (NaCl)₂, KCl, and (KCl)₂ at 1400 °C under the non-oxidative atmosphere. The feedstock contained large amounts of Ca

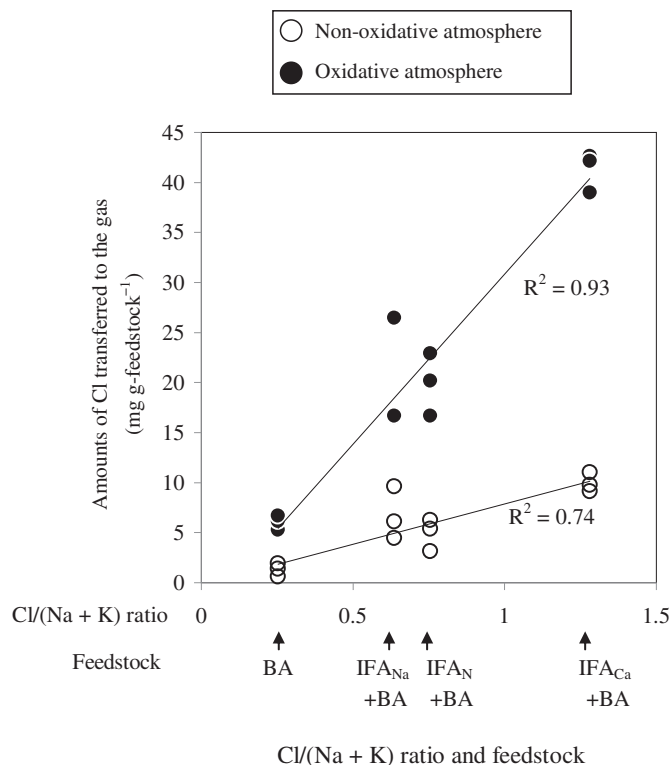


Fig. 8. Relationship between the Cl gas generations and the Cl/(Na + K) ratios. Vertical axis shows the amount of Cl transferred to the gas, and horizontal axis does the molar ratio of Cl to Na and K in the feedstock (Cl/(Na + K) ratio). The relationship between the Cl amounts and the molar ratios is described for each of the non-oxidative and oxidative atmosphere.

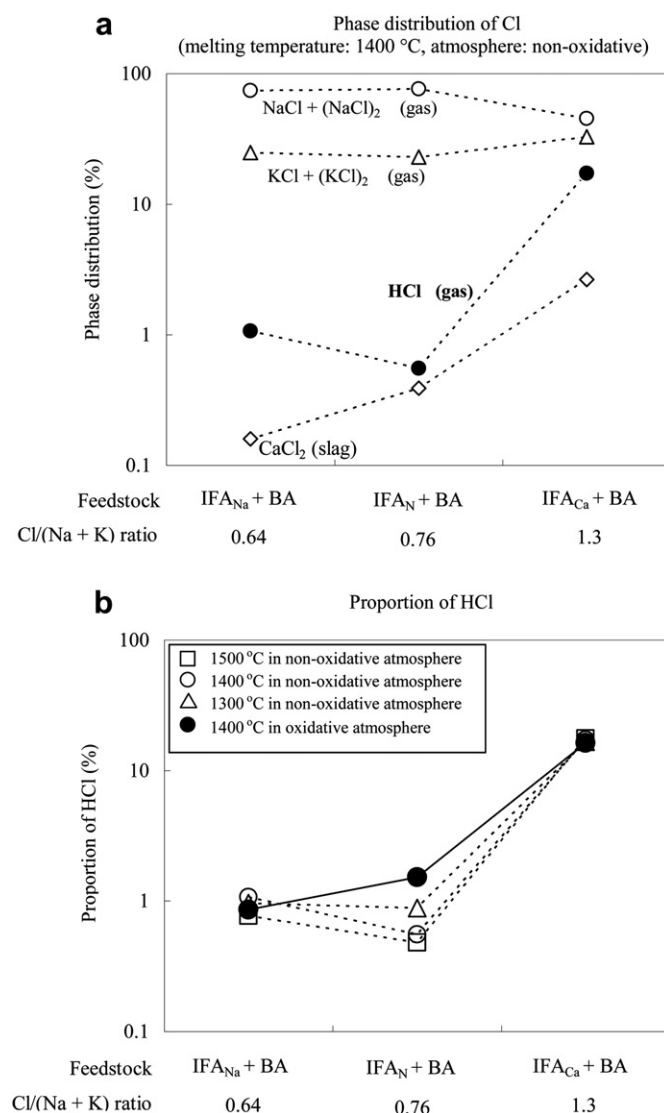


Fig. 9. Thermodynamic calculations. In Fig. 9(a), the phase distribution of Cl species between slag phase and gas phase is shown for each feedstock. In Fig. 9(b), proportion of HCl is shown for each feedstock under the different temperature and atmosphere.

Table 6

Effects of chemical compositions of IFA on the leaching characteristics of Pb from MFA.

| Chemical composition of IFA | Leaching characteristics of Pb from MFA into water | |
|-----------------------------|--|-----------------|
| | Water-soluble | Water-insoluble |
| Cl/Pb ratio | High | Low |
| Cl/(Na + K) ratio | High | Low |

(Table 3) but 99% was predicted to remain in the slag phase and not to form CaCl_2 . The proportion of HCl in the gas phase was predicted to be higher for $\text{IFA}_{\text{Ca}} + \text{BA}$ with a high $\text{Cl}/(\text{Na} + \text{K})$ ratio (>1) than for $\text{IFA}_{\text{Na}} + \text{BA}$ or $\text{IFA}_{\text{N}} + \text{BA}$ with a low $\text{Cl}/(\text{Na} + \text{K})$ ratio (<1). This trend was qualitatively consistent with the experimental results shown in Fig. 8. The effects of the atmosphere and melting temperature on the proportion of HCl are shown in Fig. 9(b). The calculation predicted that the atmosphere and melting temperature does not significantly affect the HCl generation, and the effect of the atmosphere on Cl-gas generation observed in the experiments (Fig. 8) cannot be interpreted by the equilibrium simulation.

3.2.3. Factors determining leaching characteristics of Pb in the chemical composition of IFA

To summarize the results of Sections 3.2.1 and 3.2.2, the difference in the leaching characteristics of Pb among the IFA types can be explained by the relationship among the amounts of Na, K, Cl, and Pb, as shown in Table 6. The Cl/Pb ratio in Table 6 is defined by equivalent ratios of the amount of Cl to amount of Pb in IFA. To enhance the water-leachability of Pb in MFA, a high Cl/Pb ratio is desirable for the generation of PbCl_2 with a high solubility. Furthermore, the $\text{Cl}/(\text{Na} + \text{K})$ ratio should also be high to promote Cl-gas generation, which can itself promote PbCl_2 formation. To decrease the water-leachability of Pb, on the other hand, both Cl/Pb and $\text{Cl}/(\text{Na} + \text{K})$ ratios should be low to prevent PbCl_2 formation.

4. Conclusions

This study investigated the effects of the chemical composition of IFA on the leaching characteristics of Pb from MFA generated by melting IFA. MFA generated from a real-scale melting process and lab-scale melting experiments was analyzed. In addition, theoretical behaviors of Cl that affect the leaching characteristics of Pb were simulated by a thermodynamic equilibrium calculation. The relationship among the amounts of Na, K, and Cl in IFA affected the leaching characteristics of Pb from MFA. The water-leachability of Pb in MFA can be enhanced by increasing the molar ratio of Cl to Na and K in IFA because the increase in the molar ratio promotes Cl-gas generation during the melting, which can itself promote the formation of water-soluble PbCl_2 in MFA. The relationship among the amounts of Na, K, and Cl in IFA depended on the exhaust gas

treatment systems in incinerators, and the leaching characteristics of Pb from MFA can be predicted based on the gas treatment systems. As the leaching characteristics of Pb are important for deciding between water leaching and water washing for the recovery of Pb from MFA, the findings in this study may be of value in selecting the appropriate water treatment method.

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