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Research Article

Distribution of Acid-Extractable Pb Contents in Molten Slag From Municipal Solid Waste Incineration Ash

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Abstract

In Japan, when molten slag from municipal solid waste incineration ash is used in the general environment, acid-extractable Pb contents must be lower than utilization criteria, as determined by JIS (Japanese Industrial Standard) K 0058-2 method. However, representativeness of 1,000 t of slag samples in practical usage is ineffective because only 6 g of samples are used for testing under such standards. This study has three aims: 1. to determine the variation of acid-extractable Pb contents in the 20 slag samples from one plant, 2. to estimate the probability of excess acid-extractable Pb content distributed among the 20 samples that adhere to utilization criteria, and 3. to show the importance of evaluating acid-extractable metal contents not based on a single analyzed value but probability density. In this study, twenty lots of slag were sampled from one melting plant for two months in 2006. In the first experiment, 6 g of samples were taken from each lot at random and tested. In the second experiment, 20 portions were prepared from four lots of them and 20 of 6 g samples were taken from each small portion and tested. The results indicate that significantly high Pb values derived from heterogeneous character. The dataset shows a log-normal distribution and the rates exceeding the utilization standard are discussed based on probability density function.

Keywords: Molten Slag; Pb; Distribution; Acid-Extractable Content; Log-Normal Distribution

Introduction

Approximately 200 facilities for melting municipal solid waste (MSW) or municipal solid waste incineration (MSWI) ash are running in Japan and more than 800,000 t of melting slag are generated annually [1, 2]. The molten slag is glassy and stable materials, and it is expected to be used in various civil engineering projects [3]. However, the molten slag may contain heavy metals derived from solid waste. For this reason, two test methods were conducted to determine heavy metal content on the basis of separate criteria: acid-extractable content and leaching behavior, which are outlined by JIS (Japanese Industrial Standard) K 0058-2 [4] and JIS K 0058-1 [5], respectively. Utilization criteria of molten slag for hazardous elements are shown in Table 1. Generally, with regard to MSW molten slag, the analysis and the confirmation of Pb contents is very important essential because concentration levels often exceed the utilization criteria.

Table 1. Utilization criteria of hazardous elements for molten slag in Japan

Element	Cd	Pb	Cr(VI)	As	T-Hg	Se	F	B
Acid-extractable contents (mg/kg)	150	150	250	150	15	150	4000	4000
Leaching concentration (mg/L)	0.01	0.01	0.05	0.01	0.0005	0.01	0.8	1.0

To ensure effective representation of the specimens, 50 g of samples are used for JIS K 0058-1. However, only 6 g of samples are used for JIS K 0058-2 on the contrary. Therefore the unevenness of the chemical composition of the molten slag may influence the judgment of the safety of the molten slag. In this study, the variation and distribution of the acid-extractable Pb contents in molten slag from MSWI ash are studied.

Materials and Methods

The molten slag used in this study was transported from the conveyer to the slag pit in the "A-melting plant", which is a facility for melting municipal solid waste incineration (MSWI) ash, in Japan. It is a plasma ash melting furnace system and it is one of the most popular melting processes in Japan [6-9].

Twenty lots of slag were sampled for two months in 2006; each lot was approximately 3 kg. Specimens A-1 to A-17 were generated under normal operating conditions, and specimens A-18 to A-20 were generated under trial operating conditions. The sampled molten slags were crushed to a size smaller than 2 mm using a ball mill. In the first experiment, 6 g of samples were taken from each lot at random. In the second experiment, four lots (A-2, A-9, A-18, and A-19) were selected, and the sample size was reduced by the conical quartering method to 20 portions of 50 g each. Then, 6 g samples were taken from each small portion.

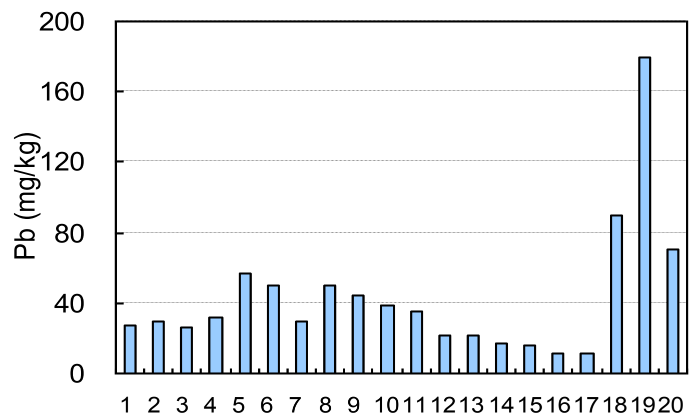
The test for acid-extractable content was performed in accordance with JIS K 0058-2. In a 500-mL polypropylene bottle, 6 g of the samples and 200 mL of 1 mol/L HCl (hydrogen chloride) were combined. The bottle was shaken at a frequency of 200 min-1 for 2 h within a range of 4–5 cm. The solution was suction-filtrated through a membrane filter with a pore size of 0.45 μm. The Pb concentration in the solution was measured by the inductively coupled plasma atomic emission spectrometer (ICP-AES; SII nanotechnology, VISTA-MPX). The same test method used for acid-extractable content was also performed for the 80 samples including 20 samples from four lots.

Results and Discussion

Variation of acid-extractable Pb content in 20 lots

Figure 1 shows the acid-extractable content of Pb from the 20 lots, the contents of which varied widely. In particular, the value of A-19 was approximately 180 mg/kg and exceeded the utilization criteria of 150 mg/kg. Specimens A-18 to A-20 contained high levels of Pb. Because this slag was generated from the melting furnace under test conditions, such high levels may be attributed to the presence of metallic Pb. Conversely, acid-extractable Pb content was significantly lower than the criteria for slag samples A-1 to A-17. However, specific variations of values were also observed. The acid-extractable Pb content varied from 12 mg/kg to 56 mg/kg, and the coefficient of variance was 45%. The factors to control the variation are probably operating condition (temperature, redox status of atmosphere, mixing of molten slag) of melting furnace, and/or Pb contents in ash. The differences of the Pb content are influenced by Pb contents in the melted waste. The ash melted in this furnace is derived from municipal solid waste incineration bottom ash, and the incinerator often incinerated shredded home electric appliances. However, the detail information of melted ash or incinerated solid waste is limited. Further research for the relationship between the molten slag and the melted incinerated ash, or the melting condition should be performed.

Figure 1. Acid-extractable Pb in the 20 lots of slag obtained for this study.



Variation of acid-extractable Pb contents in one lot

Twenty data sets of acid-extractable Pb for 4 lots of molten slag are shown in Table 2 and Figure 2. With regard to the A-2 slag, only one dataset from the 20 showed a significantly high value (S18 sample). This value was statistically judged to be outlier by the 5% significance level using the Dixon method and the values of the 19 other specimens. The detection of such sudden high value in molten slag from MSWI ash is often reported. Even melting operation of a furnace is controlled well, intrinsic uncertainty of the determined value is avoidable, because the melted incinerated ash is derived from the solid waste with various chemical composition. It is probably due to the metallic Pb derived from the incinerated solid waste, and the metallic Pb is actually detected in molten slag [10].

On the contrary, almost half of the A-19 slag samples exceeded the utilization criteria. For such specimens, mistakes in judgment of safe/dangerous of the molten slag may occur frequently, if we judge it on the basis of only one result of the compliance test. Therefore, circumspect sample preparation and/or judgement based on multiple results is necessary.

Table 2. 20 data set of acid-extractable Pb for 4 lots of molten slag.

Sample No.	A-2	A-9	A-18	A-19
S1	47	43	66	120
S2	43	39	119	306
S3	34	51	66	191
S4	38	71	88	303
S5	31	46	94	231
S6	37	40	72	264
S7	33	49	63	116
S8	38	54	63	235
S9	43	51	77	164
S10	40	43	57	100
S11	40	64	68	130
S12	37	60	68	113
S13	35	91	89	190
S14	36	45	86	144
S15	35	31	251	177
S16	34	41	86	144
S17	31	45	167	227
S18	135*	37	50	113
S19	51	37	54	138
S20	39	39	38	39

* significantly high value, and it is an outlier by Dixon's statistical test

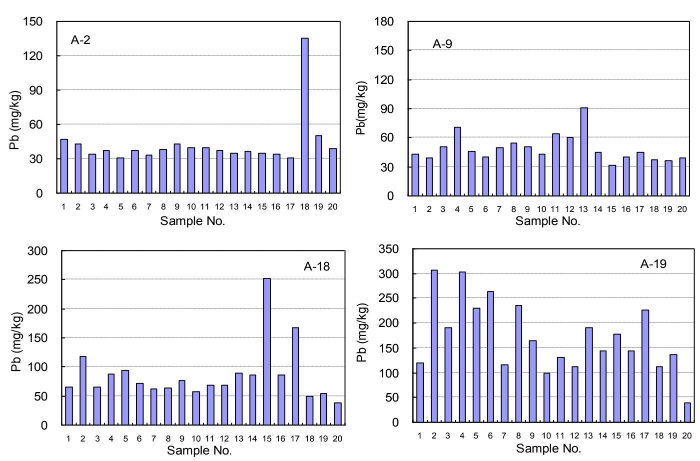
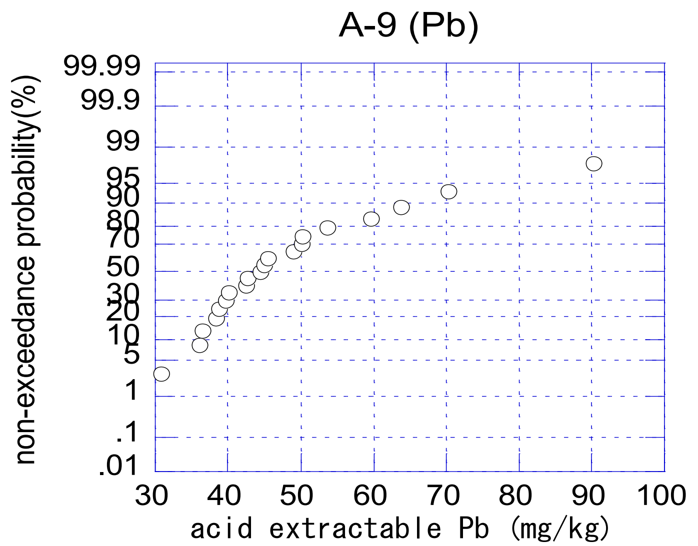


Figure 2. Acid extractable Pb in the A-2, A-9, A-18, and A-19 slags examined in this study.

In order to evaluate real risk or acid-extractable Pb contents, these datasets are analyzed statistically. The nonexceedance probabilities of logarithmic values of Pb contents for slag were plotted on a normal-probability chart. The example of A-9 slag is shown in Figure 3. Because these plots show linearity, these values can be treated as data of log-normal distribution.

Figure 3. Data plots on log-normal probability chart.



The probability-density distribution of Pb in the four selected lots of slag was estimated on the basis of the average value (μ) of logarithmic values and standard deviation (σ) in equation (1). For the sample of A-2, the outlier (S18 data) is omitted to estimate the distribution curve. The distribution curve was then drawn on the basis of the equation. The equation of probability density function is shown in Table 3 and the curve is shown in Figure 4.

Table 2. Equation of acid-extractable Pb contents distribution for 4 lots.

Lot	Equation of probability density function
A-2	$f(x)=\dfrac{1}{\sqrt{2\pi}\times 38x}\exp\left\{\dfrac{-(\log x-43)^2}{2\times(5.2)^2}\right\}$
A-9	$f(x)=\dfrac{1}{\sqrt{2\pi}\times 49x}\exp\left\{\dfrac{-(\log x-49)^2}{2\times(14)^2}\right\}$
A-18	$f(x)=\dfrac{1}{\sqrt{2\pi}\times 86x}\exp\left\{\dfrac{-(\log x-86)^2}{2\times(48)^2}\right\}$
A-19	$f(x)=\dfrac{1}{\sqrt{2\pi}\times 172x}\exp\left\{\dfrac{-(\log x-172)^2}{2\times(71)^2}\right\}$

$$f(x)=\frac{1}{\sqrt{2\pi}\sigma x}\exp\left\{\frac{-(\log x-\mu)^2}{2\sigma^2}\right\} \qquad \text{Eq. (1)}$$

The probability that the Pb contents in a slag sample exceeded the utilization standard was estimated on the basis of the distribution curve. The probability for A-2 and A-9 was nearly 0%; A-18 and A-19 values were 6.0% and ~54%, respectively. On the basis of these results, determination of safe levels in slag should rely on the concentration distribution rather than the results of only one dataset.

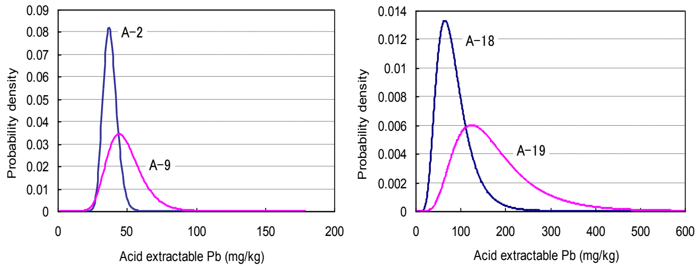


Figure 4. Estimated Pb concentration distributions in slag examined in this study.

Conclusions

The conclusions derived from this study are summarized in the following points:

Variations were observed in the amounts of acid-extractable content of Pb in the slag from the same melting plant.

Sudden high levels of acid-extractable contents of Pb were observed in a lot of MSWI ash-melting slag.

Pb concentration in MSWI ash-melting slag showed log-normal distribution in a lot.

The probability of excessive levels in samples A-2 and A-9 was nearly 0%; and that for A-18 and A-19 were 6.0% and ~54%, respectively, which illustrates the importance of evaluating the safe Pb concentration levels based on distribution.

Acknowledgements

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