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Evaluation of pretreatment methods for landfilling municipal solid waste incinerator residues

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Abstract

This study was based on the investigation of Pb, Cd, Ca, and Cl in one individual batch of bottom ash and fly ash (mixture 5:1) resulting from a specific municipal solid waste incinerator (MSWI) in Japan. This mixture ash was stabilized as the following pretreatments: water washing, carbonation, and phosphate stabilization. After the optimum processing condition for each pretreatment was determined, the performance of each pretreatment was examined using pH stat leaching tests (pH 6, 9, 12) and availability tests. For the performance tests, fly ash alone was used, instead of a mixture of MSWI residues, to observe leaching differences among the pretreatments more distinctly.

Water washing effectively removed the major elements from MSWI residues and also reduced the leachability of trace metals, such as Pb and Cd, from MSWI residues. A washing time of 15 min with an *L/S* ratio of 5 was reasonably effective. Carbonation had a significant effect on leachability in alkaline ranges (pH 9 and 12), due to carbonation only on the surface of MSWI residues, not in the center matrix. A moisture content of 10-16.7% appeared to be suitable for carbonation. Phosphate stabilization, even with small doses of phosphate (0.16 mol-PO_4^{3-} kg^{-1} sample), was very effective at reducing the leachability of heavy metals.

KEYWORDS: MSWI residues, pretreatment, water washing, carbonation, phosphate stabilization, evaluation, trace metals

1. Introduction

Incineration has played an important role in solving the problem of scarcity of available land for municipal solid waste (MSW) landfills in Japan. In 1999, approximately 78.1% of all MSW was directly incinerated, and incineration residues accounted for 68% of total landfilled MSW (Japanese White Paper on Environment 2002). In Japan, MSW incinerator (MSWI) fly ash is currently being landfilled after receiving pretreatments designated by law (Sakai 1996), while MSWI bottom ash is directly landfilled with no legally required pretreatments. Recently, the need for pretreatment of MSWI bottom ash has increased because of the high heavy metal content of bottom ash and the potential for long-term release of these toxins. However, since about 5 times more MSWI bottom ash is generated by incineration than fly ash (on a weight basis), the pretreatment method needs to be economically affordable, environmentally effective, and socially acceptable.

Several effective and economical MSWI pretreatment methods have been identified: Water washing is a simple, economical, and effective method of pretreatment, which removes most chlorides, leachable salts, and amphoteric heavy metals (Sawachi et al. 1993; Wang et al. 2001). Moreover, the leached salts have potential for use as industrial refined salts, alkalis, and acids (Horii et al. 1997). Carbonation is effective in preventing build-up of Ca in the leachate treatment systems of landfills (Noma et al. 1998) and can reduce the mobility of trace metals in alkaline fly ash (Ecke 2001; Meima et al. 2001; Reddy et al. 1994; Shimaoka et al. 1999; Seino et al. 1999). Phosphate stabilization is reported to be the most effective method for immobilizing divalent metals (Crannell et al. 2000; Eighmy et al. 1998; Furukawa et al. 1999).

However, there is little information available that evaluates the optimal processing conditions for each pretreatment or compares the three pretreatments, especially for a mixture of bottom ash and fly ash.

This study examined the optimum conditions for the three pretreatments for a mixture of bottom ash and fly ash; each pretreatment was evaluated using several leaching tests. For these tests, fly ash was used rather than a mixture of bottom and fly ash, to permit a more precise comparison of pretreatments. The results were compared with a chelating pretreatment, which is the most commonly used pretreatment in Japan. This comparison with chelating-pretreated fly ash that obtained directly from a MSWI in Japan, would provide a good criterion to evaluate the effectiveness of each pretreatment.

2. Materials and methods

2.1 Materials

Bottom ash, fly ash, and chelating-pretreated fly ash were obtained from a MSWI in city “S”, Japan. This stoker mass-burn incinerator has a processing capacity of 300 tons per day and is equipped with an electric precipitator. The flue gas is treated with an injection of dry $Ca(OH)_2$ into the gas stream. Chelating agent used in this MSWI facility consists mainly of dithiocarbamate (-NH-CSSH) group, which is widely used in Japan (Tanaka 1995). After drying at 105 ℃, bottom ash was passed through a

1.0 mm sieve. Bottom ash and fly ash were mixed with the weight ratio of 5:1 (based on actual data for landfilled waste in city “S”). This ash mixture is subsequently referred to as “B-F ash”. The chemical and physical composition of the bottom ash and fly ash are summarized in Table 1.

2.2 Methods

2.2.1 The optimal pretreatment processing conditions

A range of conditions was used for each pretreatment to determine the optimal processing conditions for each method.

2.2.1-(a) Water washing

Twenty-five grams of B-F ash were placed into three glass flasks containing 62.5, 125, and 250 mL of distilled water to reach an *L/S* of 2.5, 5 and 10, respectively. The flasks were capped and shaken horizontally at room temperature at 120 oscillations per minute for 15 min and 150 min. After shaking, liquids and solids were separated by vacuum filtration using a 1-µm glass-fiber filter. The wash water was analyzed for the elements of interest. The washed B-F ash was dried at 105℃ and then subjected to the pH 6 stat leaching test.

2.2.1-(b) Carbonation with 10% CO_2 gas

An acrylic column (internal diameter 7.1 cm, height 20 cm) was filled with 400 g of B-F ash to a depth of approximately 10 cm. Then, 10% CO_2 (90% N_2) gas was injected into the column from the bottom at a constant flow rate of 1 $Lmin^{-1}$ for 150 min. Initial moisture content (wt%) was set at 5, 10, 16.7 and 20%, on a weight basis.

The concentration of CO_2 in outlet gas, CO_2 (out), was measured every 5-10 min by GC to monitor CO_2 absorption during the carbonation process. After carbonation, the B-F ash was dried at 105℃. Finally, Ca leaching and final leachate pH were measured using Japanese leaching test No. 13 (referred to 2.2.2-(a)).

2.2.1-(c) Phosphate stabilization

Fifteen mL of phosphoric acid solution were added to 30 g of B-F ash (*L/S* 0.5). The ratio of PO_4^{3-} to B-F ash was set to 0.016, 0.16 and 0.32 mol-$PO_4^{3-}kg^{-1}$ mixed residues. Each dose was mixed for 10 min and aged for 15 min, 4 h, and 2 days in the air. Then, the pretreated B-F ash was dried at 105℃ for the subsequent pH 6 stat leaching test.

2.2.2 Leaching tests

The following tests were conducted to examine the leachability of pretreated samples versus raw samples. Each leaching test was performed in duplicate.

2.2.2-(a) Japanese leaching test No. 13 (Environment Agency of Japan 1973)

Ten grams of each sample were added to glass flasks containing 100 mL of distilled water, to reach an *L/S* of 10. The flasks were capped and shaken horizontally for 6 h at 200 oscillations per minute. After shaking, the leachate was filtered through a glass-fiber filter. The filter pore size specified in Japanese leaching test No. 13 (JLT-13) is 1 µm, but in this case a 0.45-µm membrane filter was used to permit direct comparisons with other leaching test results in this study.

2.2.2-(b) pH-stat leaching test

A pH-stat leaching test was conducted to examine the leachability of elements as a function of pH. Two grams of each sample were placed into glass flasks containing 200 mL of distilled water, to reach an *L/S* of 100. Acid or base (HNO_3 or NaOH) was added to maintain the pH at 6, 9 and 12 for 2 h. The leachate was filtered through a 0.45-µm membrane filter.

2.2.2-(c) Availability leaching test

NEN 7341 (Netherlands Normalisation Institute 1993) was used to quantify the elemental mass fraction available for leaching.

The sample is extracted twice in succession at a liquid/solid substance ratio of *L/S* = 50 Lkg^{-1}. The procedure consisted of two separate extractions: First, 2 g of the sample were placed into glass flasks containing 100 mL of distilled water (*L/S* of 50). They were stirred for 3 h (constant pH 7) and then filtered through a 0.45-µm membrane filter. Another 100 mL of distilled water were added to the remained residue (*L/S* of 50) and stirred for 3 h (constant pH 4) and then filtered through a 0.45-µm membrane filter. Each leachate was then mixed, resulting in a cumulative *L/S* of 100.

2.2.3 Analytical methods

Heavy metals in acidified filtrates (<pH2) were analyzed by GF-AAS (Hitachi Z-8200) or flame AAS (Hitachi 170-50A). The oxide composition was determined from total X-ray fluorescence (Horiba MESA-500). The concentrations of Cl and CO_2 gas were determined by ion chromatography (TOSOH CM-8) and gas chromatography (GC-TCD Hitachi 164), respectively. The pH values were reported to the nearest 0.01 unit (Horiba B212).

3. Results and discussion

3.1 The optimal pretreatment processing conditions

3.1.1 The effect of washing times and *L/S* ratios for water washing

Table 2 shows the amounts of elements that leached from the raw B-F ash as a function of washing times and *L/S* ratios. Removal rates were calculated as the ratio of the leached amount in the wash water to the total amounts of each element in the ash (Table 1). The percentages of each element that washed out of the raw B-F ash were: 67-77.4% of Cl, 0.06-0.33% of Pb, 0.03-0.05% of Ca and nearly 0% of Cd. Less Pb and Ca were leached with a washing time of 150 min than with 15 min, suggesting that precipitation and/or adsorption may occur during the longer washing time (150 min). The amounts of Pb and Ca leached increased with *L/S* ratio. Contrarily, the amount of leached Cl, a readily soluble constituent, remained nearly the same, regardless of washing time and *L/S* ratio. The leached fraction of Cd was less than 0.01% under all conditions, likely due to the very low leachability of Cd in highly alkaline solutions.

For the washed B-F ash, pH 6 stat leaching tests were conducted. This pH value was selected because the leachate pH in a landfill of alkaline MSWI residues is unlikely to decrease below 6 over a long-term period, due to the strong buffering capacity of MSWI residues (Belvi et al. 1992; Ecke 2001; Giampaolo et al. 2002; Johnson et al. 2002). The pH 6 stat leaching test results of washed B-F ash are presented in Table 3, showing the leaching of Pb, Cd, Ca and Cl as a function of the washing time and *L/S* ratio. For comparison, the pH 6 stat leaching test results of raw B-F ash are included.

Treatment efficiency was defined as follows:

$$Treatment\ Efficiency\,(\%) = \frac{BF_{raw} - BF_{treated}}{BF_{raw}} \times 100$$

where BF_{raw} = the leaching concentration from raw B-F ash in the pH 6 stat leaching test (mgL^{-1}) and $BF_{treated}$ = the leaching concentration from treated B-F ash in the pH 6 stat leaching test (mgL^{-1}).

Leaching of Cl, Pb, Cd, and Ca from washed B-F ash was reduced by 81.3-84.9%, 37.5-60.4%, 14.4-36.2% and 16.3-28.6%, respectively, compared with leaching from raw B-F ash. This suggests that water washing not only affects removal of the major elements, but also reduces the leachability of trace metals such as Pb and Cd. The significant removal of Cl by water washing may reduce the leachability of Pb and Cd, since it is well known that Cl in water increases the solubility of Pb by the formation of the Cl complexes of Pb (Cernuschi et al. 1990). The most efficient treatment for Pb, Cd and Ca was with a washing time of 15 min and *L/S* of 10, although other processing conditions had similar efficiencies. Considering the cost of subsequent leachate treatment, an *L/S* of 5 is adequate for metal reduction in comparison to an *L/S* of 10.

3.1.2 The effect of moisture content on carbonation

Fig.1 shows CO_2 (out) concentration as a function of carbonation run time. In this study, 10% CO_2 gas was used, considered the average CO_2 concentration of flue gas generated by MSWI in Japan (Tanikawa et al. 2001). Regardless of moisture content, the CO_2 (out) concentration increased exponentially with time, and leveled out at around 9%. The amount of CO_2 adsorbed by B-F ash was about 0.4-0.6 mmol

CO_2g^{-1}dry B-F ash for 150 min. The Ca content of B-F ash in this study was about 2.5 mmol Cag^{-1} dry B-F ash, which is equivalent to 1.5 mmol CaO g^{-1} dry B-F ash, considering that 60% of the total Ca content exists as CaO (Hanashima et al. 1992). This means that only one-third of CaO and/or $Ca(OH)_2$ in the B-F ash reacted with CO_2, indicating that neutralization with CO_2 may have occurred on the surface but not in the center matrix of the B-F ash. On the other hand, results of the JLT-13 tests for the treated ashes showed that the final leachate pH and leached Ca levels were almost the same in all samples. The pH ranged from 10.5-10.9, and leached Ca levels were 510-570 mgL^{-1}, suggesting that moisture content does not have a significant effect on carbonation of alkaline MSWI residues. However, at 20 % moisture content CO_2 adsorption was initially slow and unstable, suggesting that a higher moisture content may have inhibited CO_2 gas from passing through the Ca-enriched B-F ash. It has been reported that carbonation does not occur without moisture (Miyawaki et al. 1995). For the reasons mentioned above, 10-16.7% moisture content is suitable for carbonation methods.

3.1.3 The effect of PO_4^{3-} concentration and aging time on phosphate stabilization

Table 4 shows the results of the pH 6 stat leaching tests. With the exception of Cl, leaching of elements from pretreated B-F ash decreased with increased dose of PO_4^{3}. Significant reduction for Pb, 90-91%, was observed in 0.16 mol-$PO_4^{3-}kg^{-1}$ B-F ash, implying that insoluble phosphate metals such as $Pb_5(PO_4)_3Cl$ may be formed. This value is much lower than the 0.38 mol-PO_4^{3-} for 1 kg of bottom ash reported by

Crannell et al. (2000) or 1.2 mol-PO_4^{3-} for 1 kg of dry scrubber residue reported by Eighmy et al. (1997). Aging time had little effect on the leachablility of Pb, although the leaching of Cd and Ca tended to decrease slightly with aging time. As mentioned above, no significant difference in Cl leaching was observed between pretreated B-F ash and raw B-F ash. Based on these results, a dosage of 0.16 mol-$PO_4^{3-}kg^{-1}$ and a relatively short aging time (15 min) seem to be appropriate conditions for making insoluble metal phosphates.

3.2 Leaching concentration for JLT-13

The results of the JLT-13 tests are shown in Table 5. Because of high pH, high concentrations of Pb were leached from bottom ash as well as from fly ash. Based on the optimal processing condition results, B-F ash was pretreated with an *L/S* of 5 and a washing time of 15 min for water washing, 16.7% moisture content and 150 min of contact time for carbonation, and 0.16 mol-$PO_4^{3-}kg^{-1}$ mixed residues and 15 min of aging time for phosphate treatment.

The leachability of Ca from all pretreated B-F ash was reduced compared with that from raw B-F ash. This reduction was likely due to the wash-out of soluble $CaCl_2$ and $Ca(OH)_2$ by water washing, and the formation of less soluble components such as $CaCO_3$ and $Ca_5(PO_4)_3Cl$ by the carbonation process and by phosphate treatment, respectively. However, Ca reduction of chelating pretreated fly ash was relatively small compared to that of untreated fly ash. All of the pretreated materials, except the water-washed material, met Japanese standards for waste disposal of Pb (0.3

mgL^{-1}). However, it is difficult to assess the performance of each pretreatment method, due to differences in final pH. In fact, the pH of solution is known to be a major factor influencing the leachability of most metals (Stegemann et al. 1995). For better comparisons, treatment performances must be evaluated at the same pH.

3.3 Leaching behaviour of pretreated fly ash

To compare pretreatment methods, MSWI fly ash was used rather than a mixture of bottom and fly ash, because fly ash contains larger quantities of heavy metals and Cl than bottom ash. Conditions for water washing, carbonation, and phosphate stabilization were the same as those described for B-F ash in the JLT-13 test. Figure 2 shows the leaching of Pb, Cd, Ca, and Cl from each pretreated fly ash for both the pH stat test and the availability test. Fly ash pretreated by a chelating agent, the method most commonly used to stabilize fly ash in Japan, was also examined for comparison.

In comparison with the other treatments, phosphate stabilization had the greatest effect on Pb, under both acidic and alkaline conditions. Water washing effectively reduced Pb leachability over the entire pH range, which seemed to be strongly related to the significant reduction of Cl. Carbonation was effective in reducing Pb leaching at pH 12. However, as pH decreased, lead releases almost equaled that of untreated fly ash, because $PbCO_3$ and $CaCO_3$ precipitates formed by the carbonation process started to dissolve. The effect of the chelating treatment was observed in alkaline conditions (pH 9 and 12), but the leaching of Pb in chelating-preteated fly ash was similar to that of Pb in untreated fly ash in acidic conditions (pH 4 and 6), implying that insoluble chelating complexes with Pb seemed to break up in acidic conditions. These leaching phenomena of chelating-pretreated fly ash in acidic conditions corresponded to the results obtained in column tests by Sakanakura (1999).

Carbonation treatment significantly reduced the leachability of Ca in alkaline ranges (pH 9 and 12), suggesting that concentrations of dissolved Ca might be controlled by the solubility of $CaCO_3$ formed around the fly ash. However, for pH levels below 6 the results were not clearly different from those for untreated fly ash, due to the dissolution under acidic conditions of $CaCO_3$ formed on the surface of the fly ash. This suggests that, at pH levels above 6, carbonation reduces the leaching of Ca, thereby preventing clogging of the leachate collection and treatment systems of landfills. Water washing and phosphate stabilization also reduced Ca leachability, although the effects decreased as pH decreased. Chelating treatments had a negative effect on leachability over the entire pH range, except for pH 12, implying that chelating agent (dithiocarbamate (-NH-CSSH) group) seemed to have little effect on immobilization of Ca, particularly in acidic and neutral pH conditions.

All four pretreatment methods significantly reduced leachability of Cd at pH 12. In addition, phosphate stabilization greatly reduced Cd leaching at pH 9, but did not have a significantly stronger effect than the other pretreatments at pH levels below 6.

Cl, a soluble salt, was significantly reduced by water washing, but was little affected by the other pretreatments.

4. Conclusion

This study examined the optimum processing conditions for water washing, carbonation and phosphate stabilization of B-F ash and evaluated the performance of each pretreatment method by using fly ash in three kinds of batch leaching tests, which was compared with that of a chelating method. The major findings of this study are summarized here.

(1) Water washing is effective not only for removing the major elements from the ash, but also for reducing the leachability of trace metals such as Pb and Cd. However, to avoid precipitates and/or adsorption, washing time should not be too long. This method fails to meet Japanese standards for waste disposal of Pb (0.3 mg/l).

(2) Carbonation reduces the leachability of trace metals, particularly Ca, in alkaline ranges. This effect on Ca was found to be due to carbonation on the ash surface, considering the mole ratios of CO_2 adsorbed by B-F ash in comparison to the CaO and/or $Ca(OH)_2$ content of B-F ash. A moisture content of 10-16.7% is suitable for carbonation, although moisture content has little effect on carbonation unless the ash is dry**.** Carbonation may be a promising method for stabilizing metals in MSWI residues, provided that the leachate pH does not decrease below 6.

(3) Phosphate stabilization has an excellent effect, reducing the leachability of heavy metals over the entire pH range, particularly for Pb. A dose of 0.16 mol-$PO_4^{3-}$$kg^{-1}$ sample is sufficient. Aging time has little effect on the release of metals.

5. References

1. Belevi, H., Stampfli D.M. & Baccini, P. (1992) Chemical behaviour of municipal solid waste incinerator bottom ash in monofills, Waste Management & Research **10**, 153-167.

2. Cernuschi, S., Giugliano, M. & De Paoli, I. (1990) Leaching of residues from MSW Incineration. Waste Management & Research **8**, 419-427.

3. Crannell BS, Eighmy TT, Krzanowski JE, Eusden Jr. JD, Shaw EL & Francis CA (2000) Heavy metal stabilization in municipal solid waste combustion bottom ash using soluble phosphate, Waste Management **20,** 135-148.

4. Ecke, H (2001) Carbonation for Fixation of Metals in Municipal Solid Waste Incineration (MSWI) Fly Ash, Doctoral Thesis, 1-20.

5. Environment Agency of Japan, Japanese Environment Agency Notification No. 13, 1973 (in Japanese)

6. Furukawa, K., Uchibo, A. Uchida, T., Itou, I., Yoshida, T., Kambayashi, H., Abe, S. & Kokado, M. (1999) Leaching behavior of untreated and phosphate pretreated fly ash from municipal solid waste melting facilities and minerals formed in phosphate-treated fly ash, Waste Management Research (in Japanese), **10(5),** 276-283.

7. Giampaolo, C., Lo Mastro, S., Polettini, A., Pomi, R. & Sirini, P. (2002) Acid neutralization capacity and hydration behaviour of incineration bottom ash – Portland cement mixtures, Cement and Concrete Research, **32,** 769-775.

8. Hanashima N, Simaoka T, Higuchi S, Yamaguchi R、Hori Y, & Nagaoka H. (1991) Control of inorganic salts by piling up the wastes (in Japanese). Proceedings of the 2nd Annual Conference of the Japan Society of Waste Management Experts (in Japanese), 281-284.

9. Horii Y., Higuchi, S., Shimaoka, T. & Hanashima, M. (1997) The treatment technology of landfill leachates containing condensed inorganic salts, Waste Management Research (in Japanese), **8(7),** 529-539.

10. Johnson, C. A. & Gerhard Furrer (2002) Influence of biodegradation process on the duration of CaCO3 as a pH buffer in municipal solid waste incinerator bottom ash, Environmental Science and Technology, **36(6),** 215-220.

11. Kuen-Shen Wang, Kung-Yun Chiang, & Kae-Long Sun (2001) Effects of a water-extraction process on heavy metal behavior in municipal solid waste incinerator fly ash, Hydrometallurgy, **62**, 73-81.

12. Ministry of the Environment, Japanese White Paper on the Environment 2002

13. Miyawaki, K., Matsuto, T., Tanaka, N., Matsuo, T. & Masuda, T. (1995) Neutralization of leachate pH by carbon dioxide absorption in incineration ash landfill. Proceedings of Environmental Engineering Research, **32,** 417-423.

14. NEN 7341, Leaching characteristics of building and solid waste materials-leaching tests-determination of the availability of inorganic components for leaching. Draft 1993 (previously part of NVN 2508), Netherlands Normalisation Institute, The Netherlands, 1993.

15. Noma, Y. and Kida, A. (1997) Carbonation treatment for highly alkaline

incinerator fly ash and its leaching characteristics, Waste Management Research (in Japanese), **8(4),** 129-137.

16. Reddy, K.J., Gloss, S.P. & Wang, L. (1994) Reaction of CO_2 with alkaline solid wastes to reduce contaminant mobility, Water Research, **28(6),** 1377-1382.
17. Research Group for Wash Out Waste (WOW) System (2001) Guidebook for Leachate Treatment Technology (in Japanese), 151-190.
18. Sakai, S. (1996) Municipal solid waste management in Japan. Waste Management **16**, 395-405
19. Sakanakura, H. (1999) Study on stabilization of heavy metal in municipal solid waste incinerator residues by chelating agents (in Japanese). PhD thesis, 53-65
20. Sawachi, M., Minoda, T. & Fukunaga, I. (1993) Water washing method for MSW incinerator fly ash treatment (in Japanese). Journal of Japan Waste Management Association **46 (193),** 148-153.
21. Seino, A., Kim, S.Y., Tanaka, N. & Matsuto, T. (1999) Study on controlling trace metals from bottom ashes by carbonation treatment (in Japanese). Proceedings of the 10th Annual Conference of the Japan Society of Waste Management Experts. 497 ‐ 499.
22. Shimaoka, T., Miyawaki, K., Hanashima, M., Itoh, I., Yoshid, T. & Uchida, T. (1999) Insolubilization of Pb in fly ash using the exhaust gas from incineration plant. Proceedings of the Seventh International Waste Management and Landfill Symposium, CISA, Environmental Sanitary Engineering Centre, Cagliari, Italy, S. Margherita di Pula, Cagliari, Italy, 565-572.
23. Stegemann, J.A., Schneider, J., Baetz, B.W. & Murphy, K.L. (1995) Lysimeter

washing of MSW incinerator bottom ash. Waste Management & Research **13**, 149 – 165.

24. Tanaka Y. (1995) Recent performance on the treatment of heavy metals by chelating methods (in Japanese). Sangyo to Kankyo **24(9)**, 77–79.
25. Tanikawa, N., Oikawa, T., Masuko, T. & Urano, K. (2001) An estimation method for CO_2 emissions from municipal waste incinerators by continuous O_2 analyzer, Waste Management Research (in Japanese), **12(3),** 131-136.

Table 1. Chemical and physical composition of the bottom and fly ash used in the study

Element	Concentration[a] mean ±SD[b]		Method
	Bottom ash	Fly ash	
Si (%)	14.5±1.22	8.2±1.51	FAAS
Ca (%)	10.1±0.23	12.8±0.26	FAAS
Al (%)	5.4±1.48	5.0±1.35	FAAS
Fe (%)	2.5±0.18	0.7±0.01	FAAS
Na (%)	1.6±0.04	5.1±0.09	FAAS
K (%)	1.2±0.02	4.1±0.51	FAAS
Cl (%)	0.89±0.01	12.4±0.02	IC
Cu (mg/kg)	9700±140	1130±13	GF-AAS
Mg (mg/kg)	8350±240	10300±180	GF-AAS
Zn (mg/kg)	2290±50	8100±110	GF-AAS
Pb (mg/kg)	1240±25	2810±15	GF-AAS
Mn (mg/kg)	547±17	430±12	GF-AAS
Cr (mg/kg)	208±46	126±5.30	GF-AAS
Cd (mg/kg)	3.2±0.52	102±0.50	GF-AAS
Oxide composition			
SiO_2 (% dry wt.)	38.5±0.68	13.1±0.25	XRF
CaO (% dry wt.)	26.2±0.26	38.0±0.43	XRF
Al_2O_3 (% dry wt.)	13.6±0.29	4.9±0.23	XRF
Fe_2O_3 (% dry wt.)	6.3±0.07	2.1±0.04	XRF
K_2O (% dry wt.)	3.3±0.06	8.3±0.14	XRF
Physical propety			
Water content (%)	31.3±0.7	0.1±0.01	
Ignition loss[c] (%)	7.7±0.21	3.9±0.28	

[a] HNO_3/HCl total digestion/ Flame AAS or Graphite Furnace AAS

[b] SD: standard deviation (n=2)

[c] Ignition Loss at 600℃ for 3 hr

IC: ion chromatography

XRF: X-ray fluorescence

Table2. Amount of elements leached in the wash water

			Pb		Cd		Ca		Cl	
Washing time (min)	*L/S* ratio (mL/g)	pH of leachate	Leached content[a] ±SD[b] (mg/kg)	Removal[c] (%)	Leached content ±SD (mg/kg)	Removal (%)	Leached content ±SD (mg/kg)	Removal (%)	Leached content ±SD (mg/kg)	Removal (w/w %)
15	2.5	12.4	2.23±0.32	0.14	0.002	0.01	3460±280	0.03	24700±1050	77.4
	5	12.4	3.71±0.19	0.24	0.002	0.01	4260±186	0.04	22400±678	70
	10	12.3	5.1±0.07	0.33	0.002	0.01	5780±325	0.05	21400±1360	67
150	2.5	12.3	0.91±0.23	0.06	ND[d]	0	2770±196	0.03	24700±980	77.4
	5	12.3	1.38±0.09	0.09	ND	0	3560±330	0.03	21500±1500	67.3
	10	12.1	1.45±0.15	0.09	0.001	0	4870±180	0.05	22100±1260	69.2

Concentration in leachate (mg/l) = [a]leached content (mg/kg)/(*L/S*-ratio)

[b] SD: standard deviation (n=2)

[c] Removal = leached content in water (mg/g) /(total content (mg/g)) × 100

[d] ND stands for not detectable

Table 3. pH 6 stat leaching results of washed B-F ash by washing time and L/S ratio

		Pb		Cd		Ca		Cl	
Washing time (min)	*L/S* ratio (mL/g)	Concn[a] ±SD[b] (mg/L)	Treatment Efficiency[c] (%)	Concn ±SD (mg/L)	Treatment efficiency (%)	Concn ±SD (mg/L)	Treatment efficiency (%)	Concn ±SD (mg/L)	Treatment efficiency (%)
15	2.5	0.038±0.003	40.6	0.026±0.004	25.3	567±35	18.8	55±3	83.4
	5	0.034±0.001	47	0.027±0.006	21.3	534±36	23.6	50±5	84.9
	10	0.025±0.001	60.4	0.022±0.002	36.2	499±32	28.6	55	83.4
150	2.5	0.042±0.009	34.3	0.030±0.003	14.4	570±40	18.4	62±3	81.3
	5	0.037±0.006	41.4	0.027±0.001	21.7	549±47	21.4	53	84
	10	0.040±0.003	37.5	0.030±0.002	14.4	585±35	16.3	59±3	82.2
Raw B-F ash		0.063±0.005	-	0.034±0.003	-	699±33	-	331±2	-
Detection limit		0.002		0.001		1	-	5	-

Leached content (mg/kg) = [a] concentration in leachate (mg/l)×100 (L/S ratios of pH 6 stat leaching tests)

[b] SD: standard deviation (n=2)

[c] Treatment efficiency = (the leaching concn of raw B-F ash - the leaching concn of washed B-F ash) / the leaching concn of raw B-F ash × 100

Table 4. pH 6 stat leaching results of pretreated B-F by PO_4^{3-} concentration and aging time

Amount of PO_4^{3-} (mol PO_4^{3-} /kg B-F ash)	Aging time (hour)	Pb Concn[a] ±SD[b] (mg/L)	Pb Treatment efficiency[c] (%)	Cd Concn ±SD (mg/L)	Cd Treatment efficiency (%)	Ca Concn ±SD (mg/L)	Ca Treatment efficiency (%)	Cl Concn ±SD (mg/L)	Cl Treatment efficiency (%)
0.016	0.25	0.036±0.005	42.7	0.030±0.001	13	486±14	30.4	324±9	2
	4	0.039±0.001	37.9	0.030±0.001	11.7	466±6	33.3	304±5	8
	48	0.040±0.004	35.8	0.028±0.002	17	466±10	33.3	322±11	2.7
0.16	0.25	0.006±0.002	90.5	0.025±0.004	26.9	403±6	42.4	322±6	2.6
	4	0.006±0.005	91.1	0.025±0.004	25.2	400±3	42.8	334±1	-1.1
	48	0.006±0.001	91.1	0.022±0.003	34.2	370±15	47.1	335±6	-1.3
0.32	0.25	0.003±0.002	95.7	0.021±0.003	37.2	326±14	53.3	320±9	3.3
	4	0.004±0.003	94.2	0.02±0.001	41.4	339±7	51.5	311±10	6
	48	0.004±0.001	94.1	0.016±0.002	52.2	270±15	61.3	331±9	0
Raw B-F ash		0.063±0.005	-	0.034±0.003	-	699±33	-	331±2	-
Detection limit		0.002		0.001		1	-	5	-

Leached content (mg/kg) = [a] concentration in leachate (mg/l)×100 (L/S ratios of pH 6 stat leaching tests)

[b] SD: standard deviation (n=2)

[c] Treatment efficiency = (the leaching concn of raw B-F ash - the leaching concn of pretreated B-F ash) / the leaching concn of raw B-F ash × 100

Table 5. pH and concentrations of elements in the leachate after JLT-13 test (units (except pH) : mg/l)

Sample	Pb Concn+SD[a]	Cd Concn+SD	Ca Concn+SD	Cl Concn+SD	pH of leachate
Bottom ash	3.03±0.42	ND[b]	603±25	544±6	12.4
Fly ash	44.30±2.70	0.004	3120±21	13113±110	12.9
Chelating pretreated fly ash	0.21±0.01	ND	2730±14	12353±137	11.8
Water washed B-F ash	1.2±0.05	ND	463±26	327±3.5	12.0
Carbonated B-F ash	0.01	ND	510±23	2436±53	10.5
Phosphate stabilized B-F ash	0.03	ND	692±15	2142±14	11.5
Raw B-F ash	4.66±0.32	ND	921±12	2209±71	12.4
Japanese standard for waste disposal	0.3	0.3	-	-	-
Detection limit	0.002	0.001	1	5	

[a] SD: standard deviation (n=2)

[b] ND stands for not detectable

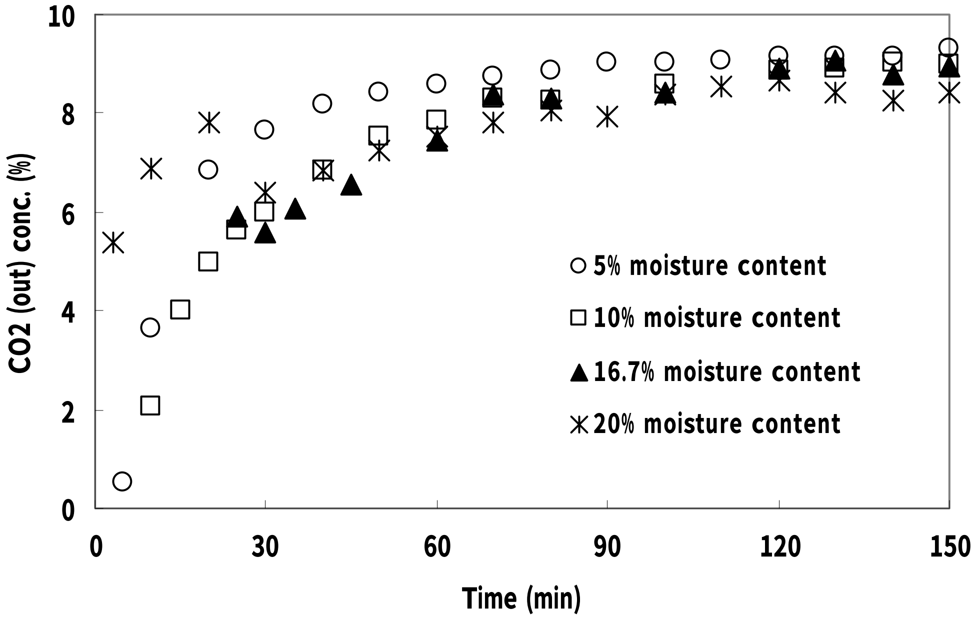

Fig. 1 CO_2 (out) with time

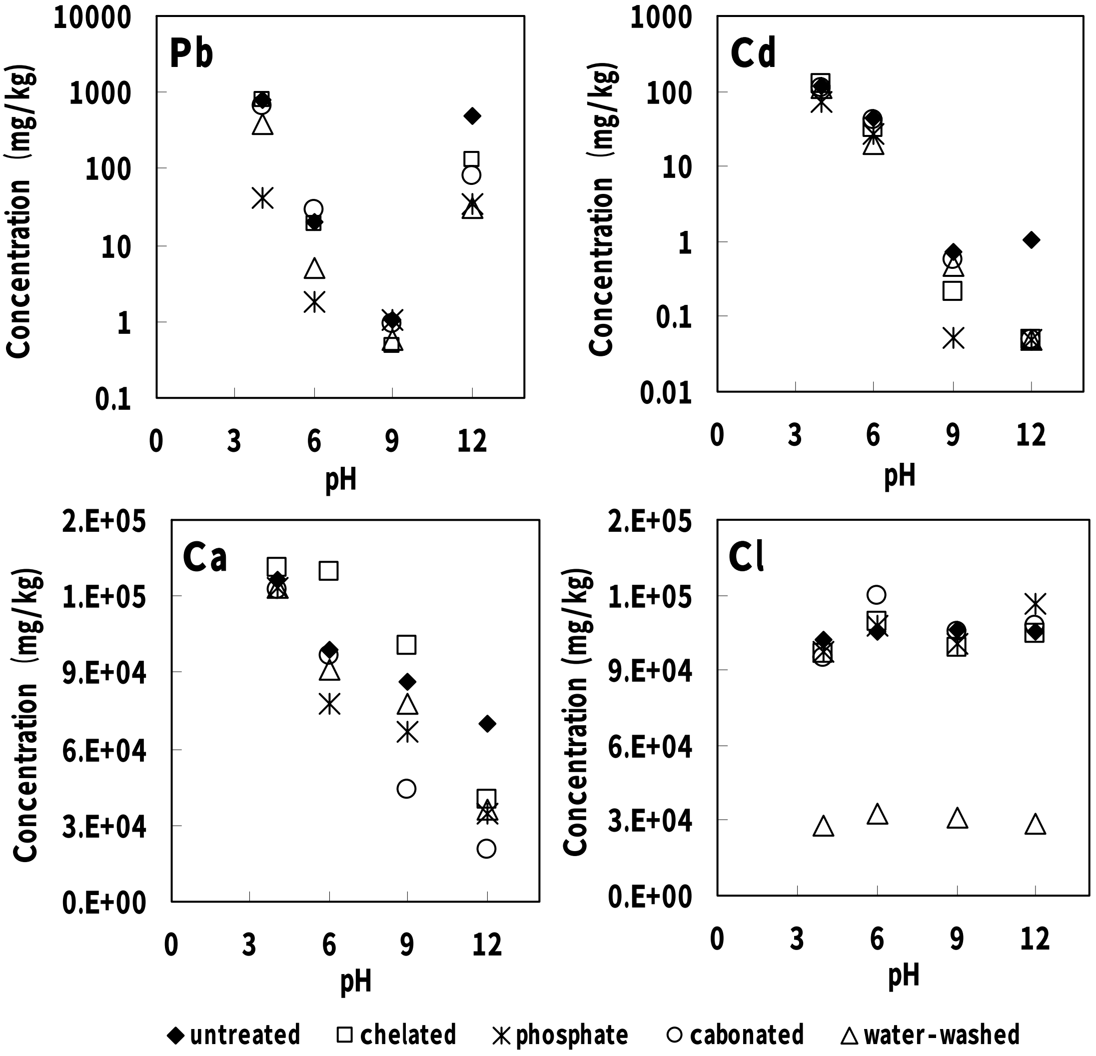

Fig 2. The leached content of Pb, Cd, Ca, and Cl from pH stat leaching test and availability test. (Due to the higher amount of heavy metals and Cl in fly ash, fly ash was used instead of a mixture of bottom ash and fly)