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Extraction of heavy metals from MSW incinerator fly ashes by chelating agents

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Abstract

An extraction process has been studied on a laboratory scale for the pretreatment of municipal solid waste (MSW) incinerator fly ash to remobilize Cr, Cu, Pb, and Zn. Five different types of fly ashes were treated with HCl, nitrilotriacetic acid (NTA), ethylenediaminetetraacetate (EDTA), or diethylenetriaminepentaacetate (DTPA) in a batch process in the pH range 2.5–10. The extraction of heavy metals by HCl was dependent on pH, increasing with increasing acid concentration. The efficiency of the chelating agents was independent of pH. By the treatment with 3.0% EDTA or DTPA, 20–50% of Cr, 60–95% of Cu, 60–100% of Pb, and 50–100% of Zn were extracted in the pH range 3–9. NTA was also effective in extracting Cr, Cu, and Zn. The maximum extraction of Cr, Cu, Pb, and Zn was obtained at 0.3–1.0% concentration of the chelating agents. NTA was effective in extracting Pb at a concentration as low as 0.1%. Extraction behavior of other elements during the treatment was also studied. The leaching test on the residues after the treatment with chelating agents showed that the fly ashes were successfully detoxified to meet the guideline for landfilling. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Chelating agent; Acid treatment; Detoxification; Extraction; Fly ash; Heavy metals

1. Introduction

With diversification and increase of the amount of municipal solid waste (MSW), the incineration process has been increasingly adopted by local authorities as an important

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waste management technology, by which the volume of waste can be reduced to 10–20%. In Japan, as much as 75.5% of 5.05×10^7 metric tons MSW was treated by the incineration method in 1994 [1]. One of the serious problems accompanying this process is the generation of fly ash, which contains various hazardous elements such as lead, chromium, cadmium, arsenic, antimony, etc. These elements in fly ashes are in the more soluble forms than those in bottom ashes [2]. These elements are derived mainly from household waste of small sealed lead battery, nickel–cadmium battery, copper, chromium, arsenic (CCA)-preserved wood, and antimony-containing flame-proofed products. By the incineration process, household wastes were changed to fly ash in a transfer ratio of 33% of lead, 92% of cadmium, and 45% of antimony [3]. In Japan, MSW incinerator fly ash is classified as a specially controlled general waste for which one of the specified pretreatment processes is required prior to landfilling. The Waste Disposal and Public Cleansing Law [4] specifies the following four pretreatment methods to prevent the release of hazardous elements from fly ash: (1) vitrification [5]; (2) cement solidification [6,7]; (3) chemical treatment [8]; and (4) extraction with acid or other solvents [9]. Vitrification is the most reliable process by which heavy metals are stabilized and the fly ash volume can be reduced to 1/4–1/6, but its disadvantage is relatively high cost of treatment and construction. Cement solidification has been the most popular method because of its simple operation and cost effectiveness. But it has been reported that the stability of heavy metals in fly ash solidified by cement may be reduced by water-soluble sulfate and organic acid and carbon dioxide gas produced by the decomposition of organic substances in the MSW [10]. In addition, cement solidification is not suited for Pb-rich fly ashes, since Pb tends to leach out from the solid phase at high pH, which is caused by cement itself. Chemical treatment has been also a popular method because of its easy operation. Heavy metals may leach out from chemically treated fly ashes due to change in chemical environment such as pH [10].

From the environmental and economical point of view, the extraction process seems attractive since fly ash can be detoxified by remobilizing hazardous elements from the solid phase. For this purpose, an acid or a chelating agent has been used. An excess

Table 1
Complex formation constants of aminopolycarboxylic acids (20°C, IS = 0.1) [11]

Metal	NTA	EDTA	DTPA
Cu	12.96	18.8	21.55
Pb	11.39	18.04	18.8
Zn	10.67	16.5	18.4
Fe (III)	15.9	25.1	28
Mg	5.41	8.79	9.3
Al	11.4 ^a	16.3	18.6
K	na	0.8	na
Na	1.22	1.66	na
Ca	6.41	10.69	10.83

na: not available.

^a25°C, IS = 0.1.

amount of acid is generally required to leach out heavy metals, which results in a problem of handling of acidic residue. One of the advantages of chelating agents is that they generally work under moderate pH conditions. Among various chelating agents, aminopolycarboxylates such as nitrilotriacetate (NTA), ethylenediaminetetraacetate (EDTA), and diethylenetriaminepentaacetate (DTPA) form stable water-soluble complexes with various metal ions as shown in Table 1 [11]. It can be expected to leach out heavy metals from fly ash by treating with such chelating agents. On the other hand, chelating agents containing thiol (–SH) or dithiocarbamate (–NH–CSSH) group have been used [12] for the abovementioned (three) chemical treatment to stabilize fly ash by forming insoluble chelate complexes with heavy metals.

The objective of the present study was to evaluate, on a laboratory scale, the performance of NTA, EDTA, and DTPA to detoxify MSW incinerator fly ash for subsequent safe landfill disposal or utilization, which was compared with that of acid treatment. In addition, extraction of elements other than heavy metals during the treatment was studied to better understand the extraction process, since chelating agents can form complexes also with Al, Ca, Mg, etc., which are abundant in fly ashes.

2. Materials and methods

2.1. Sampling and characterization of fly ashes

Five different types of fly ashes were collected from MSW incineration plants. The fly ash samples were sieved through a 0.59-mm standard sieve and the fraction smaller than 0.59 mm in diameter was subjected to the subsequent experiment. To determine the elemental composition, a fly ash was digested with $\text{HNO}_3 + \text{HCl}$ (1 + 3) at boiling temperature [13]. The digested liquid was filtered with 0.45 μm nitrocellulose membrane filter and the filtrate was analyzed for Cr, Cu, Pb, Zn, Al, Ca, Fe, Mg, and S by inductively coupled plasma (ICP) spectrometry (ICPS 7500; Shimadzu, Kyoto, Japan). Na and K were determined by emission spectrometry (AA6200; Shimadzu, Kyoto, Japan). The content of C, Cl, and Si was determined by X-ray fluorescent spectroscopy (SXF-1200; Shimadzu, Kyoto, Japan) without acid digestion. The characteristic pH of a fly ash was obtained by suspending 0.5 g of fly ash in 25 ml of deionized water. The surface area of a fly ash was measured by the BET method (Flow Sorb II 2300; Micromeritics, Norcross, GA, USA).

2.2. Extraction of heavy metals

An amount of 0.5 g of each fly ash was placed in a series of polycarbonate centrifuge tubes and approximately 20 ml of deionized water or 3.75% chelating agent aqueous solution was added. A 3 M HCl was added so as to bring the pH in the range 2.5–10. The sample volume was made up to 25 ml by adding deionized water, so that the final chelating agents concentration became 3.0%. The suspensions were shaken for 24 h in a

20°C thermostat, then centrifuged with a refrigerated centrifuge. The supernatant was filtered with a 0.45- μm nitrocellulose membrane filter. The filtrate was analyzed for Cl^- and SO_4^{2-} by ion chromatography (LC-6A; Shimadzu, Kyoto, Japan). The heavy metals concentrations were determined by the ICP method. Hexavalent Cr was determined by the colorimetric method [14]. As chelating agents, nitrilotriacetic acid (NTA), ethylenediaminetetraacetic acid (EDTA), and diethylenetriaminepentaacetic acid (DTPA) of reagent grade (Kanto Chemical, Osaka, Japan) were used without further purification.

To study the effects of chelating agent concentration, 0.1%, 0.3%, 0.6%, 1.0%, and 3.0% solutions were used in the pH range of 6.0–6.5. Thereafter, the same procedure was followed.

The residues obtained from the chelating agents treatment were subjected to the leaching test specified in Notification No. 13 of the Environment Agency, Japan [15]. An amount of 50 g of fly ash was mixed with 500 ml of water (pH 5.8–6.3). The suspension was shaken for 6 h at room temperature and filtered with a glass fiber filter paper. The filtrate was analyzed for the heavy metals in the same way as above.

3. Results and discussion

3.1. Characterization of MSW incinerator fly ashes

General information about the five MSW incinerator fly ashes is given in Table 2. In every case, lime was used for the treatment of acid flue gas such as HCl and SO_x . Due to residual lime, the equilibrium pHs of the fly ash suspensions (solid-to-water ratio of 1:50) were in the pH range of 11.7–12.1. As their physical characteristics, they were composed of fine particles and the surface areas were generally low.

The elemental compositions of the five fly ashes are shown in Fig. 1. Approximate values are given for the contents of oxygen, which are calculated as the remainder from the total contents of the measured elements. The chemical characteristics of the fly ashes varied substantially, depending on the furnace type, flue gas treatment, and operating conditions, as well as with the MSW feed composition. The major elements in the fly

Table 2
Samples of MSW incinerator fly ash

Fly ash	Furnace	Dust collector	Fraction < 0.59 mm (%)	Surface area (m^2/g)	pH ^a
A	fluidized bed	fabric filter	100	2.24	11.7
B	stoker	electrostatic precipitator	93.9	3.24	12.1
C	stoker	fabric filter	90.8	3.74	12.1
D	stoker	fabric filter	95.9	5.57	12.1
E	stoker	fabric filter	94.9	4.71	12.1

^aEquilibrium pH of a suspension (1 + 50).

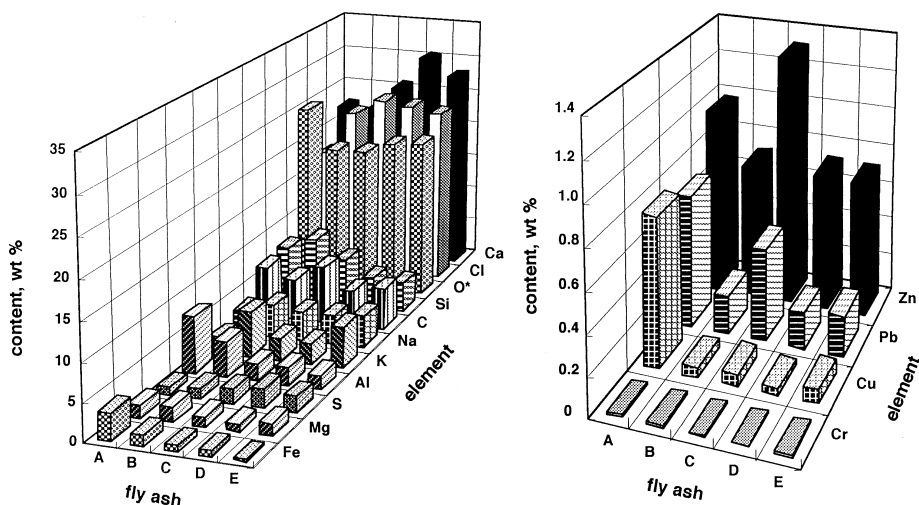


Fig. 1. Elemental composition of MSW incinerator fly ashes (* Approximate value by calculation).

ashes were Ca, Cl, O > Si, C, Na, K, Al > S, Mg, and Fe. As the minor elements, hazardous heavy metals such as Zn, Pb, Cu, and Cr were detected below 1%. Further, other heavy metals such as Ni, Sb, Se, Cd, and Co were also contained in the fly ashes. The content of these metals were < 0.01% by weight percent. Fly ash A was the most contaminated sample in terms of contents of heavy metals. During MSW incineration, oxides of Si, Al, Fe, and other metals form the core of the fly ash particles, and then volatile species such as Pb and Cd chlorides condense on the surface [16,17]. It is estimated that the metals are in the form of oxide, carbonate, chloride and sulfate. Most of the Ca, Cl, Na, and K are in the form of soluble salt. A significant amount of carbonated paper residue was observed especially in the fractions > 0.59 mm.

3.2. Extraction of heavy metals from fly ash

The effectiveness of NTA, EDTA and DTPA in extracting Cr, Cu, Pb, and Zn from the fly ashes are collectively shown in Figs. 2–5, respectively. The results are expressed in terms of percentage of extracted heavy metals to their original content and compared with the results of acid treatment (control). Since the fly ashes were highly alkaline, 3 M HCl was added to all the samples so that the final pH became < 10. Therefore, the term “acid treatment” is employed in this study to refer to the control experiment.

Chromium was the least amount of minor element present in the fly ash samples. Acid treatment of the fly ash was not effective in extracting Cr in the final pH range of > 5, with only 20–30% of Cr being extracted at pH around 4 (Fig. 2). Although more Cr was extracted by the three chelating agents, their percentage extraction was as low as 20–45% in the final pH range of 3–9, which was almost independent of pH. Such low percentage extraction of Cr can be attributed to the fact that the greater proportions of Cr

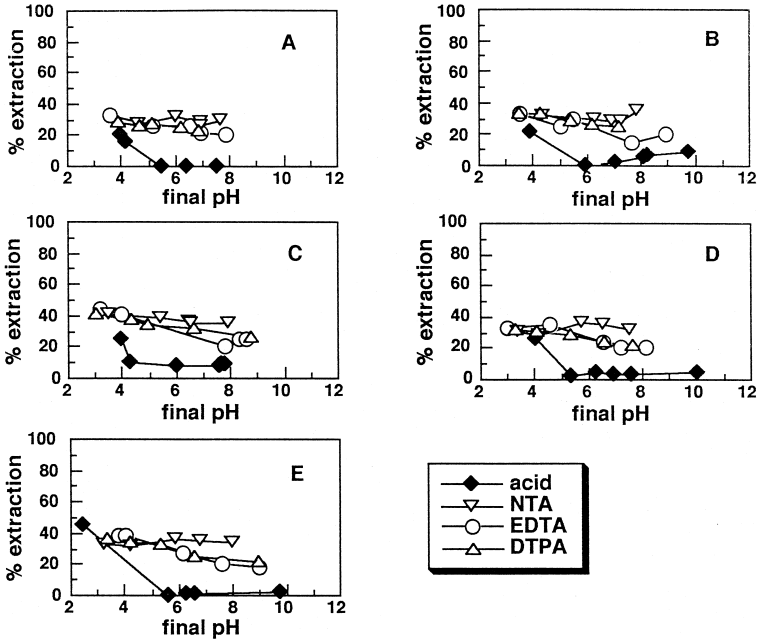


Fig. 2. Extraction of Cr from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

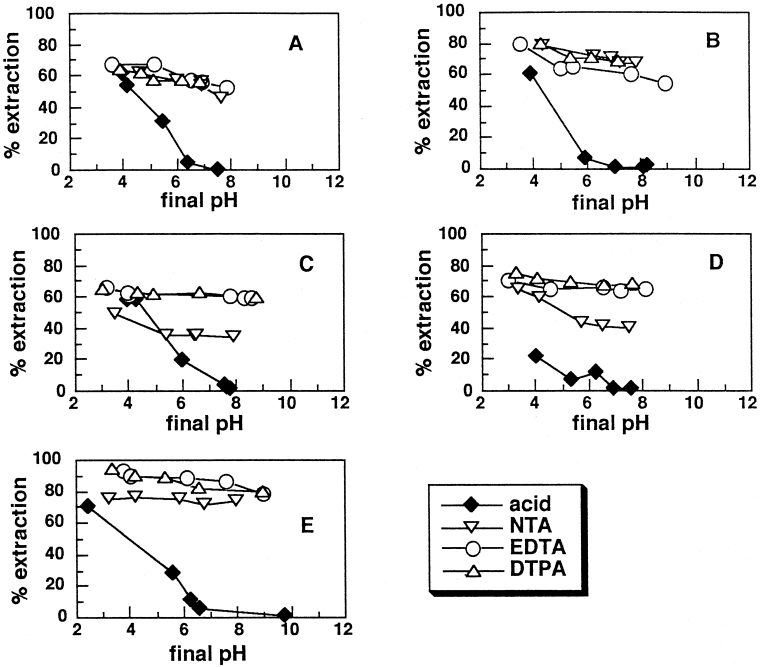


Fig. 3. Extraction of Cu from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

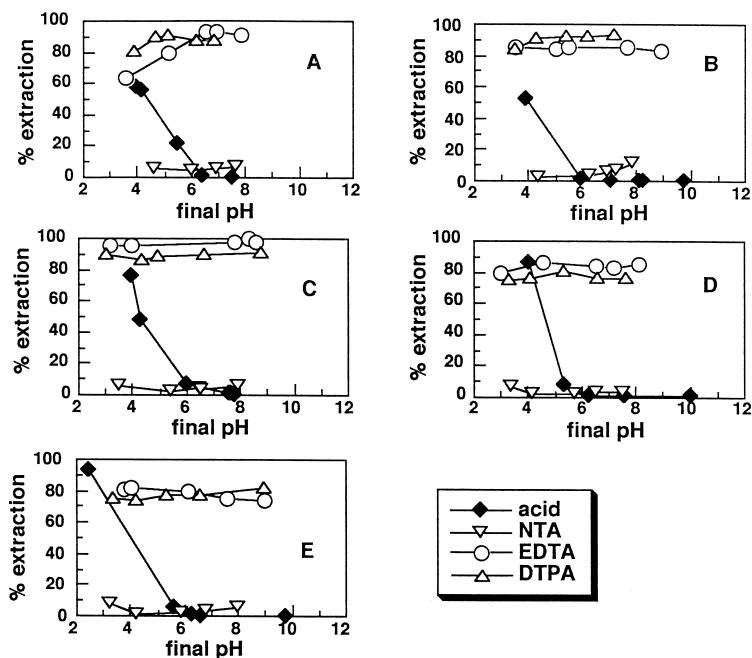


Fig. 4. Extraction of Pb from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

exist in the non-extractable form in fly ashes [18]. NTA showed a slightly higher percentage of Cr extraction in the neutral to alkaline pH range than EDTA and DTPA.

The percent of Cu extraction by acid treatment increased with decreasing pH, attaining more than 60% extraction at final pH 4 or less except fly ash D (Fig. 3). At a final pH of around 8, little Cu was extracted by acid. EDTA and DTPA gave high percentages of Cu extraction in the pH range of 3–9, which was slightly affected by the pH. NTA was also effective in extracting Cu in a wide pH range but the extraction percentage was lower than those for fly ashes C, D, and E. Thus, more Cu was extracted by the chelating agents at neutral pH range than by acid.

Treatment of the fly ashes with acid at final pH < 4 attained 50% or a higher percentage of Pb extraction (Fig. 4). The result of fly ash E shows that Pb can be extracted almost completely by acid treatment at pH around 2, which required the addition of large amounts of acid. Little Pb was extracted at final pH > 6 by acid treatment. Both EDTA and DTPA were effective in extracting Pb in the pH range of 3–9 attaining 70–100%, extraction which was almost independent of pH. In contrast to EDTA and DTPA, NTA was entirely ineffective in extracting Pb. This result can be attributed to an electrostatic re-adsorption of $\text{Pb}(\text{NTA})_2^{4-}$ onto positively charged oxide such as Al_2O_3 as proposed by Elliot and Brown [19]. They reported that 1:2 Pb–NTA complex is formed in the presence of excess NTA and that high ionic strength reduces Pb extraction. As discussed in Section 3.3, a high concentration of NTA (3.0%) was employed in this study.

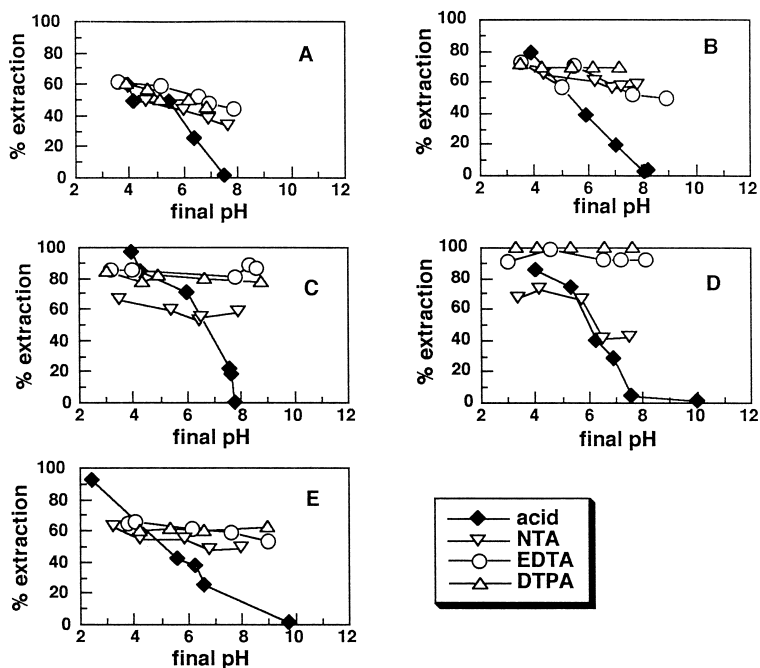


Fig. 5. Extraction of Zn from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

Zinc was a heavy metal evenly distributed in the fly ashes. The extraction curves of Zn by acid treatment shifted to the alkaline side, indicating that Zn was more soluble than the other heavy metals. EDTA and DTPA showed 80–100% Zn extraction from fly ashes C and D, but NTA gave a lower extraction. For fly ashes A, B, and E, the three chelating agents attained similar results.

In general, the efficiency of the acid treatment was dependent on final pH, increasing with an increasing acid concentration. The acid treatment was as effective as the chelating agents in extracting the heavy metals in the low pH range, which resulted in a problem of handling of acidic residues. On the other hand, the efficiencies of the chelating agents on the four heavy metals were almost pH-independent, on which the degree of the complex formation constants was not reflected. By the treatment with 3.0% EDTA or DTPA, 20–50% of Cr, 60–95% of Cu, 60–100% of Pb, and 50–100% of Zn were extracted in the pH range of 3–9. NTA was also effective in extracting Cr, Cu, and Zn. Practically, it is advantageous that extraction can be performed in a neutral pH range. The efficiencies varied with the type of fly ashes probably due to difference in the forms of heavy metals and effects of the major elements such as Ca, Cl, and sulfate.

3.3. Concentration effects of chelating agents

The effects of concentration of the chelating agents on the extraction of Cr, Cu, Pb, and Zn from fly ash A were studied at 0–3.0% concentrations and pH 6.0–6.5 with a

solid-to-liquid ratio of 1:50. Fly ash A was selected because of its high contents of heavy metals in comparison with the other fly ashes. The results are shown in Fig. 6. As a whole, NTA, EDTA, and DTPA were effective in solubilizing Cr, Cu, Pb, and Zn from the fly ash. In the tested pH range of 6.0–6.5, little heavy metals except Zn were extracted by the acid treatment.

The extraction of Cr increased with increasing concentrations of DTPA and NTA, the maximum percentage of extraction being attained at the concentration of 0.6% or higher. But the concentration effect of EDTA was smaller than those of DTPA and NTA, attaining maximum extraction at 1.0%. For Cu, there were little differences in the concentration effects among the chelating agents. The percent Cu extraction reached the maximum at 0.6%. Lead showed different results from those of the other metals. The maximum Pb extraction was attained at 0.3–1.0% concentrations of EDTA and DTPA. At 3.0% concentration, the Pb extraction slightly decreased. The extraction of Pb by NTA was maximum at 0.1% concentration attaining 75% extraction, decreasing significantly with increasing NTA concentration, which indicates that NTA is effective at low concentrations. The negative effect may be due to formation of $\text{Pb}(\text{NTA})_2^{4-}$, which is reabsorbed on the positively charged solid. Due to such negative effect, NTA showed extremely low percentage of Pb extraction as shown in Fig. 4, where as much as 3.0% NTA was added. The concentration effects of the three chelating agents were not significant for the extraction of Zn attaining the maximum at 1.0% concentration. Zinc in the fly ash was soluble in the neutral pH range and dissolved to some extent by the acid treatment, as shown in Fig. 5.

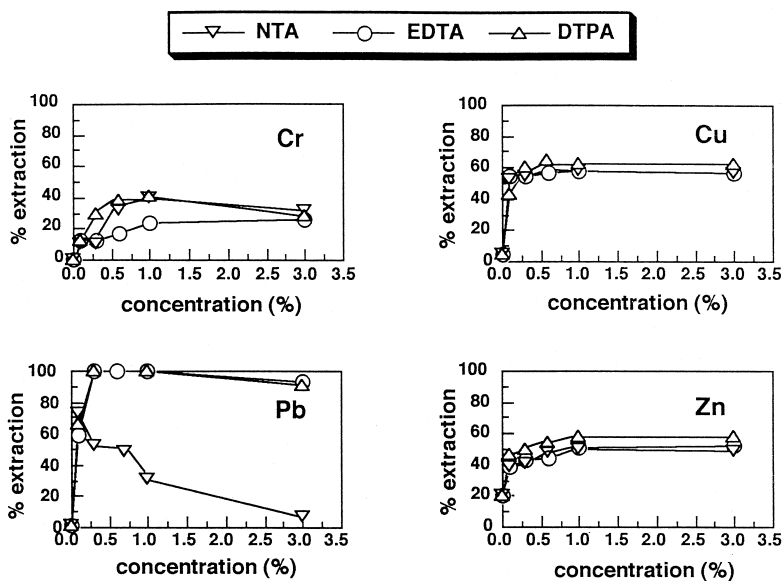


Fig. 6. Effects of chelating agents concentration on the extraction of heavy metals from fly ash A (pH 6.0–6.5).

3.4. Extraction of other elements from MSW incinerator fly ashes

As discussed in the preceding sections, treatment of fly ashes with the chelating agents were generally effective in facilitating the extraction of Cr, Cu, Pb, and Zn. Extraction of the major elements during the treatment with acid or the chelating agents is discussed in this section. Sodium and potassium salts in fly ashes are highly soluble in water in the wide pH range from 2 to 12 [2]. Calcium, magnesium, chloride, and sulfate salts exist also in the soluble forms, which potentially could result in undesirable effects such as scaling and corrosion if fly ashes are not properly managed.

Calcium is one of the most abundant elements of the fly ashes since excess lime is injected to the flue gas to neutralize acids. According to the X-ray photoelectron spectrometric study on fly ashes [2], Ca exists in the form of CaSO_4 and $\text{CaCl}(\text{OH})$ or CaCl_2 . Fig. 7 shows that Ca salts in the fly ashes were highly soluble and the treatment of the fly ashes with EDTA or DTPA gave similar extraction patterns as those of acid treatment attaining 70–90% extraction in the wide pH range. Such little difference in the Ca extraction between the acid and EDTA or DTPA treatment is due to highly soluble nature of Ca salts in the fly ashes. On the other hand, NTA treatment drastically decreased the Ca extraction for every fly ash. This result is probably due to the same phenomenon of readsorption of $\text{Ca}(\text{NTA})^-$ or $\text{Ca}(\text{NTA})_2^{4-}$ complex on positively charged solid particles.

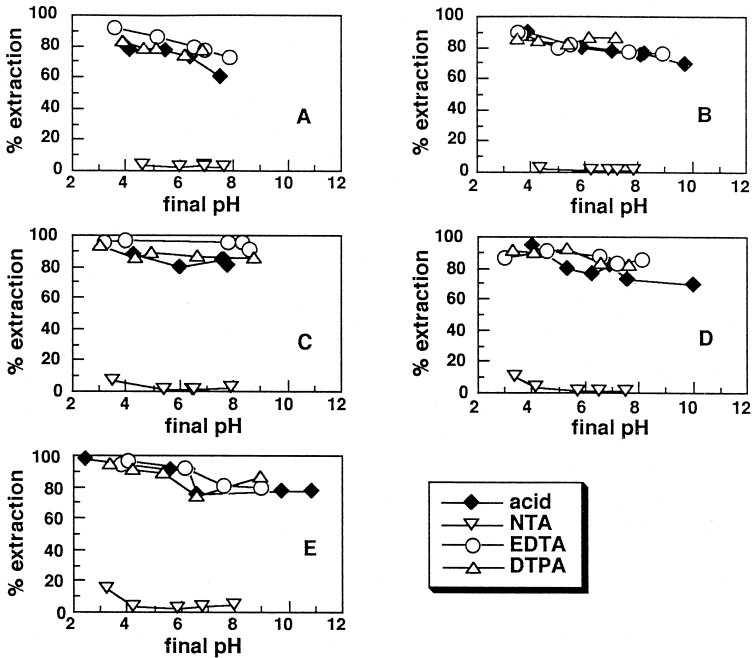


Fig. 7. Extraction of Ca from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

There were little difference in the extraction of Mg among the treatment with the chelating agents or acid (Fig. 8). But the average percent of Mg extraction changed among the fly ashes. Similarly, the extraction of chloride ions was almost identical among the fly ashes as well as among the extractants except acid treatment of fly ash B and EDTA treatment of fly ash E, which gave lower percentage extraction of chloride (Fig. 9).

Fig. 10 shows that NTA remarkably facilitated the extraction of sulfate from the fly ashes, attaining 100% sulfate extraction. This may be related to the lower extraction of Pb and Ca. There were appreciable difference in the sulfate extraction among the chelating agents as well as the fly ashes .

The extraction of Al by both chelating agents and acid was pH-dependent, increasing with decreasing pH and becoming insignificant at neutral pH range (Fig. 11). The treatment with NTA and EDTA facilitated extraction of Al than acid treatment in the pH range of 4–6.

Fig. 12 shows that Fe was highly resistant to acid treatment except at pH around 2. Although the chelating agents form Fe(III) complexes of high stability constants as shown in Table 1, little Fe was extracted by the chelating agents in the pH range of 4–9 except fly ash E. Similarly, Si was highly resistant to acid treatment in the neutral-to-alkaline pH range (Fig. 13). Little Si was extracted by the treatment with NTA, EDTA, or DTPA in the pH range of 3–9. It should be noted that the three chelating agents

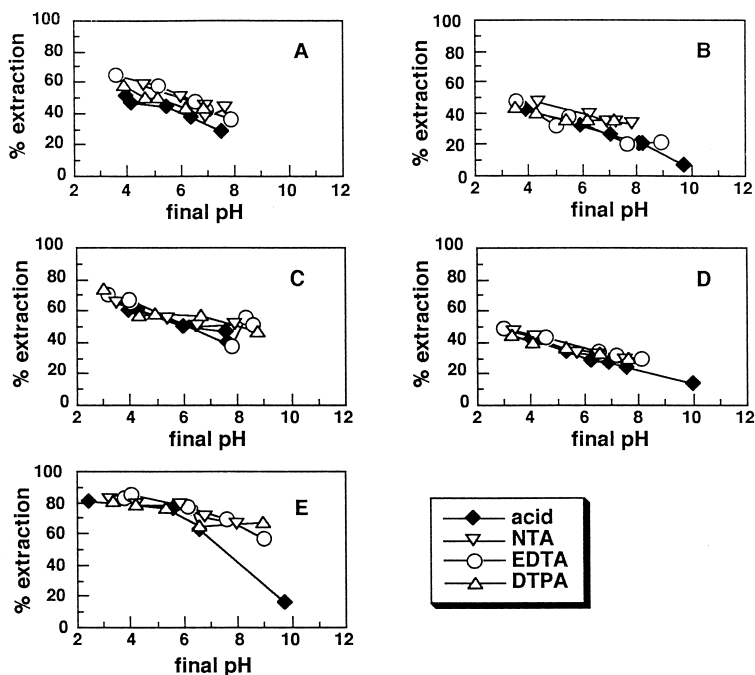


Fig. 8. Extraction of Mg from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

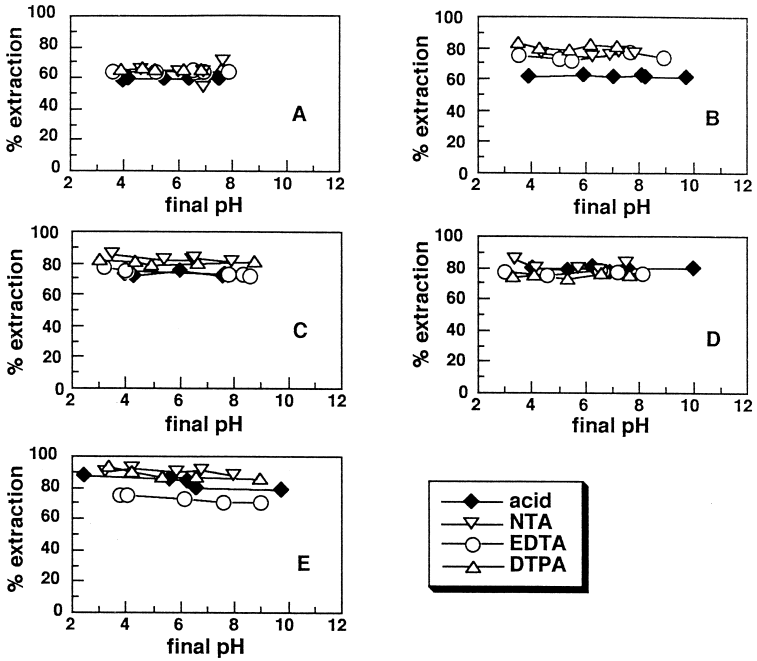


Fig. 9. Extraction of Cl from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

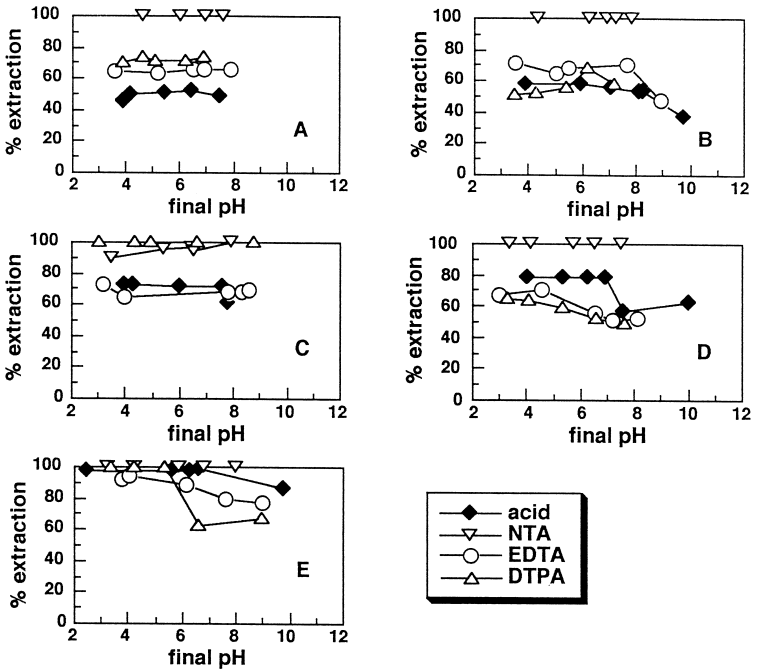


Fig. 10. Extraction of SO₄ from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

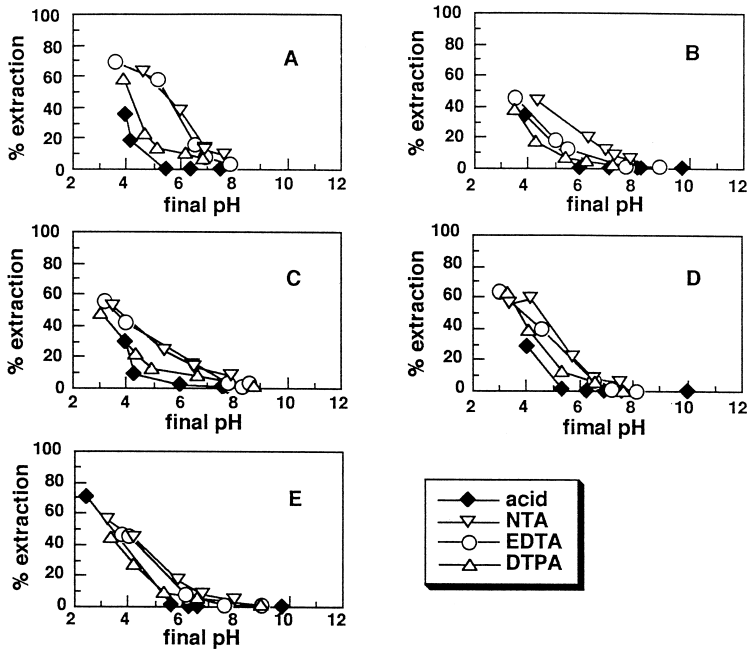


Fig. 11. Extraction of Al from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

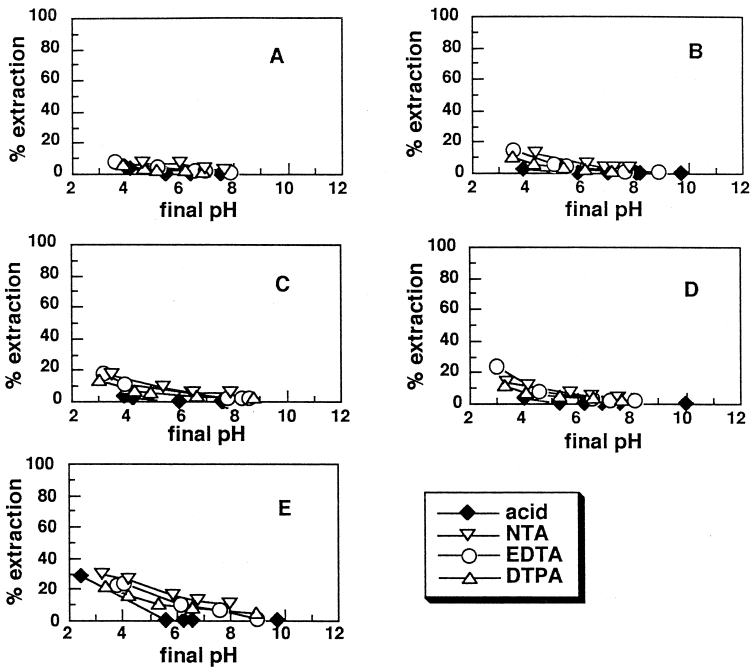


Fig. 12. Extraction of Fe from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

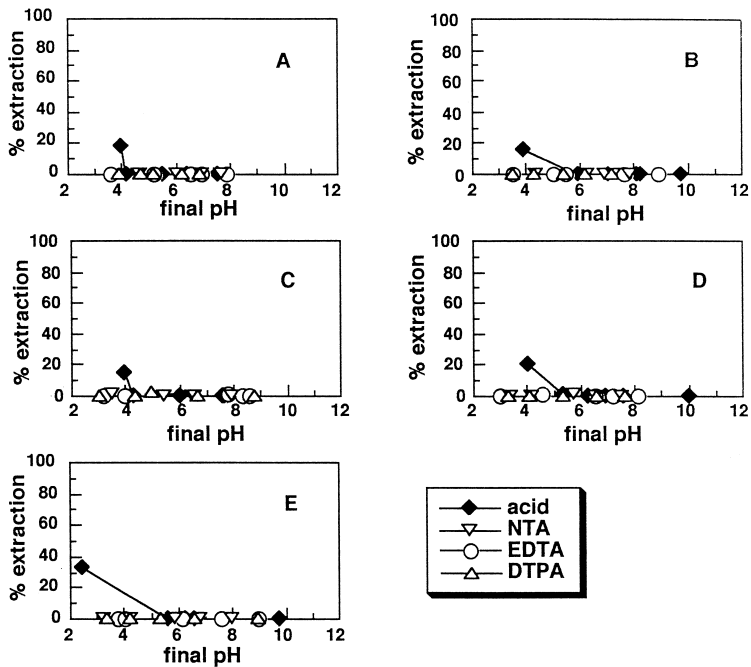


Fig. 13. Extraction of Si from the fly ashes (acid = HCl, concentration of chelating agents = 3%).

operated to depress leaching of Si at $\text{pH} < 4$, whereas an appreciable amount of Si was dissolved by the acid treatment, which created difficulty of dewatering. Therefore, Si and Fe are in the stable form in fly ashes.

3.5. Leaching test

In Japan, fly ash is finally disposed of either in a controlled or an isolated-type landfill site. The latter accepts fly ashes of leaching potential of hazardous elements. The landfilling type is decided by the leaching test specified by the Environment Agency [15]. In this study, the residues recovered from the treatment of fly ash A–E with chelating agents at $\text{pH} 3.84\text{--}5.12$ were examined by this leaching test. The results are shown in Table 3. The concentration of metals leached was compared with the guideline values [20]. Only 0.004 mg/l Pb dissolved from the residue of fly ash A treated with EDTA and undetectable Pb from all the other residues. From 0.058 to 0.062 mg/l of hexavalent Cr was detected in all the leachate from the fly ash residues, which was sufficiently below the guideline value of 1.5 mg/l . But it should be noted that Cr has the potential to leach out even after the treatment with the chelating agents. Only from fly ash A residue was leaching of Cu detected. The Cu concentrations in the leachate from the other residues were undetectable. Similarly, appreciable amounts of Zn dissolved from residues B and D. As a whole, the concentrations of heavy metals in the leachate were much lower than the guideline values. Therefore, the treatment of the fly

Table 3

Results of the leaching test for the fly ash residues treated with chelating agents

Fly ash	Extractant	Leachate pH	Heavy metal content (mg/l)			
			Cu	Cr (VI)	Pb	Zn
A	NTA	4.65	0.250	0.066	nd	nd
	EDTA	4.13	0.200	0.062	0.004	nd
	DTPA	3.91	0.032	0.059	nd	nd
B	NTA	4.44	nd	0.066	nd	0.168
	EDTA	4.09	nd	0.063	nd	0.164
	DTPA	3.84	nd	0.060	nd	0.153
C	NTA	3.54	nd	0.062	nd	nd
	EDTA	4.09	nd	0.060	nd	nd
	DTPA	4.14	nd	0.058	nd	nd
D	NTA	4.21	nd	0.062	nd	0.058
	EDTA	4.77	nd	0.062	nd	0.056
	DTPA	4.37	nd	0.060	nd	0.052
E	NTA	4.84	nd	0.060	nd	nd
	EDTA	5.12	nd	0.061	nd	nd
	DTPA	4.57	nd	0.058	nd	nd
Guideline value			^a	1.5	0.3	^a

nd: not detected.

^aGuideline is not set.

ashes with chelating agents was effective in detoxifying fly ashes to meet the landfilling guideline.

4. Conclusions

An extraction process has been studied on a laboratory scale for the pretreatment of MSW incinerator fly ash to remobilize Cr, Cu, Pb, and Zn. Five different types of fly ashes were treated with HCl, NTA, EDTA, or DTPA in a batch process in the pH range of 2.5–10. The chemical characterization showed that the fly ashes were composed of Ca, Cl, O > Si, C, Na, K, Al > S, Mg, and Fe. As the minor elements, hazardous heavy metals such as Zn, Pb, Cu, and Cr were detected. The extraction of heavy metals by HCl treatment was dependent on pH, increasing with increasing acid concentration. The efficiency of the chelating agents was almost independent of pH. By the treatment with 3.0% EDTA or DTPA, 20–50% of Cr, 60–95% of Cu, 60–100% of Pb, and 50–100% of Zn were extracted in the pH range of 3–9. NTA was also effective in extracting Cr, Cu, and Zn but gave negative effect on the extraction of Pb probably due to readsorption of Pb(NTA)_2^{4-} complex. The maximum extractions of Cr, Cu, Pb, and Zn were obtained at 0.3–1.0% concentration of the chelating agents. The extraction of Pb by NTA was maximum at 0.1% concentration attaining 75% extraction, decreasing significantly with increasing NTA concentration.

Calcium salts in the fly ashes were highly soluble in the wide pH range, 70–90% being extracted by acid, EDTA, and DTPA. Magnesium, chloride, and sulfate were also

highly soluble, being extracted by acid and the chelating agents. Extraction of sulfate was facilitated by NTA. Extraction of Al was pH-dependent and facilitated by the chelating agents. Iron and Si were stable against extraction with acid and the chelating agents. The leaching test on the residues after the treatment with chelating agents showed that the fly ashes were successfully detoxified to meet the landfilling guideline.

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References

- [1] Ministry of Health and Welfare, Japan, Generation and treatment of municipal solid wastes in 1994, Sangyo to Kankyo 26 (1997) 90–93, No. 11 (in Japanese).
- [2] A. Kida, Y. Noma, T. Imada, Chemical speciation and leaching properties of elements in municipal incinerator ashes, Waste Management 16 (1996) 527–536.
- [3] K. Nakamura, S. Kinoshita, H. Takatsuki, The origin and behavior of lead, cadmium and antimony in MSW incinerator, Waste Management 16 (1996) 509–517.
- [4] Ministry of Health and Welfare, Japan, Notification No. 194 of the Ministry of Health and Welfare, July 3, 1992, in Japanese.
- [5] C. De Labarre, Vitrification of incineration wastes, Verre (Versailles) 3 (4) (1997) 33–39.
- [6] J.R. Conner, Chemical Fixation and Solidification of Hazardous Wastes, Van Nostrand-Reinhold, New York, 1990.
- [7] S. Macakova, M.T. Hepworth, N. Pliesovska, J. Hatala, F. Siaka, Immobilization of heavy metals from MSW incinerator ash via use of sored cement, J. Solid Waste Technol. Manage. 24 (1) (1997) 27–36.
- [8] T. Takahashi and M. Kamikita, Methods for stabilizing heavy metals in fly ashes from incinerator, Jpn. Kokai Tokkyo Koho Jpn. pp. 9, 248, 542 [97, 248, 542].
- [9] H. Katsuura, T. Inoue, M. Hiraoka, S. Sakai, Full-scale plant study on fly ash treatment by the acid extraction process, Waste Management 16 (1996) 491–499.
- [10] T. Shimaoka, M. Hamashima, Behavior of stabilized fly ashes in solid waste landfills, Waste Management 16 (1996) 545–554.
- [11] A.E. Martell, R.M. Smith, Critical Stability Constants, Amino Acids Vol. 1 Plenum, New York, USA, 1976.
- [12] Y. Tanaka, Recent performance on the treatment of heavy metals by chelating methods, Sangyo to Kankyo 24 (1995) 77–79, No. 9 (in Japanese).
- [13] S. Tokunaga, T. Hakuta, Analysis of chemical compositions of fly ashes by X-ray fluorescence spectroscopy, 8th Conf. Jpn. Soc. Waste Manage. Expert (1997) 422–424.
- [14] Standard Methods for the Examination of Water and Wastewater, in: A.D. Eaton, L.S. Clesceri, A.E. Greenberg (Eds.), 19th edn., APAAH, AWWA, WEF, Washington, DC, 1995, pp. 3–59.
- [15] Environment Agency, Japan, Testing methods for metals in industrial wastes, Notification No. 13 of the Environment Agency, February 17, 1973, in Japanese.
- [16] R.R. Greenburg, W.H. Zoller, G.E. Gorder, Composition and size distributions of particles released in refuse incineration, Environ. Sci. Technol. 12 (1978) 566–573.
- [17] J.W. Graydon, D.W. Kirk, Characterization of fly ash from a municipal solid waste incinerator, in: EPD Congress 1992, TMS, 1992, pp. 327–350.

- [18] L.C. Tan, V. Choa, J.H. Tay, The influence of pH on mobility of heavy metals from municipal solid waste incinerator fly ash, *Environ. Monit. Assess.* 44 (1997) 275–284.
- [19] H.A. Elliott, G.A. Brown, Comparative evaluation of NTA and EDTA for extractive decontamination of Pb-polluted soils, *Water, Air, Soil Pollut.* 45 (1989) 361–369.
- [20] Prime Minister's Office, Japan, Ordinance No. 5 of the Prime Minister's Office, February 17, 1973, in Japanese.