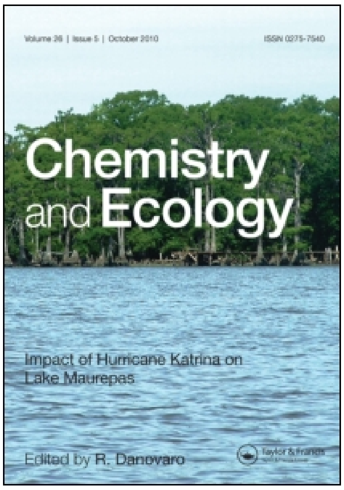

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Leaching Characteristics of Lead and Cadmium From Waste-To-Energy Residues in Sea Water

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LEACHING CHARACTERISTICS OF LEAD AND CADMIUM FROM WASTE-TO-ENERGY RESIDUES IN SEA WATER

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Laboratory studies were conducted to determine leaching characteristics of waste-to-energy (WTE) residues in sea water to provide initial evaluation on the potential impact of WTE residues on the marine environment. Both loose and stabilized WTE residues were investigated for the leachability of lead (Pb) and cadmium (Cd) in sea water. Results of the study showed that loose WTE fly ash was very reactive in sea water; release of Pb and Cd occurred immediately (<0.5 hr) after the ash came in contact with sea water. Stabilization (treated with Portland cement and other additives), however, significantly reduced leaching of Cd and Pb. While ocean disposal of untreated WTE residues would not be advisable, utilization of stabilized WTE residues for beneficial uses should be investigated.

KEY WORDS: Incineration ash, leaching, Pb, Cd, ocean disposal

INTRODUCTION

Incineration of municipal solid waste (MSW) may result in the generation of potentially harmful substances which may be found in the stack emission or in the ash (Neal and Schubel, 1987; Frame, 1988) generated by a waste-to-energy (WTE) facility. WTE residue is the term for the solid waste that remains when the waste is burned in the incinerator. Generally at a mass-burn facility (i.e., MSW is burned without separation prior to combustion), 90% of the ash residue is bottom ash, an aggregate-like material with a large range in particle size (0.01–2.5cm); the remaining 10% is fly ash, a finely divided material of uniform particle size (<0.5mm). Fly ash and bottom ash may be combined or handled separately at the incinerator facility. If a scrubber system (e.g., flue gas desulphurization) is employed to remove stack gases emitted by the incinerator, additional solid residues (e.g., gypsum, anhydrite, lime, limestone) will be produced.

Two major problems encountered in the management of WTE ash residues are: (1) the production of large quantities of the ash residues and (2) the enrichment of contaminants in the ash residues. Production of MSW incineration ash will continue to increase because more MSW incinerators are being built to solve the problem of managing the increasing quantities of MSW due to rapid population growth. It is estimated that 19 million tons of WTE ash alone will be generated in the United States by the year 2000 (National Research Council, 1988). It is well known that the ash is enriched in trace metals and contaminants of environmental concern (Greenberg *et al.*, 1978; Law and Gordon, 1979; Fraser and Lum, 1983; Denison, 1988, U.S. EPA, 1990) and that there are significant variations in elemental concentration and composition (U.S. EPA, 1990; Yoo, 1991). Further, the physical and chemical properties of WTE ash residue vary with source of MSW being burned and operational procedures used at individual incinerator facilities (Sawell and Constable, 1988; Stewart, 1989).

Coastal states in the United States and other maritime countries, including islands with limited space for waste disposal (Shieh and Duedall, 1991), are increasingly turning to incineration for the management of the voluminous amounts of MSW being produced. Because maritime areas are close to or surrounded by the ocean, improper management of WTE ash residues may easily lead to marine pollution. Therefore, environmentally acceptable methodologies for ash management, including ash utilization, must be developed.

Currently, the practice of ash dispersal is mainly landfills. However, under some circumstances, the landfilling of ash is becoming unfavoured because of environmental, political, or cost considerations. Thus, technology and/or methodology involving ash recycling and utilization in a safe form, such as ash-concrete, is being developed (Wiles, 1989; Shieh *et al.*, 1990). Stabilization/solidification of the ash to form ash-concrete is currently being considered a major approach for the use of the ash residue. Marine application of stabilized ash-concrete using coal fly ash, for example, as a material for the construction of artificial reef at sea has been investigated (Woodhead *et al.*, 1985; Sheih *et al.*, 1990; Collins, *et al.*, 1990).

In this paper, laboratory studies conducted to evaluate metal leaching from both loose and stabilized WTE ash are presented. The interaction of untreated WTE ash residue with sea water and the effectiveness of stabilization on reducing metal leachings from WTE ash in sea water are discussed. The information from this study will be useful to decision makers who are managing the WTE ash wastes.

MATERIAL AND METHODS

Material

Loose WTE ash residues used in this study were fly ash, scrubber residue, and bottom ash collected from a mass-burn WTE facility. The elemental composition of the ash residues is shown in Table 1.

Stabilized ash-concretes used in the study contained either 100 parts bottom ash (mix BA), 70 parts bottom ash/30 parts scrubber residue (mix BASR), or 60 parts bottom ash/40 parts fly ash (mix BAFA). The methodology of stabilization was described by Jonsson (1990). In general, loose WTE ash residue was mixed with known amounts of cement and water followed by curing of the mixture at a constant temperature and humidity over a specified period of time to form the stabilized ash-concrete product. The composition of the mix designs of the stabilized ash-concretes used in the study are shown in Table 2.

Sea Water Leaching Test on Loose WTE Ash

A systematic series of leaching experiments were conducted on the loose WTE ash to determine its leaching of Pb and Cd in sea water. Three replicate ash samples were placed in polypropylene plastic bottles containing sea water to form a solid/liquid

Table 1 Concentrations of selected elements in waste-to-energy ash residues

Element	*Fly Ash*	*Bottom Ash*	*Scrubber Residue*
Si (%)	15±1	22±1	19±1
Ca (%)	6.8±0.3	7.7±0.9	8.6±0.7
Fe (%)	1.1±0.1	6.8±0.1	3.4±0.1
Al (%)	4.6±0.2	2.8±0.1	6.5±0.9
Mg (%)	1.10±0.06	0.90±0.09	1.80±0.07
Zn (%)	4.2±0.2	0.40±0.04	0.40±0.03
Pb ($\mu g\, g^{-1}$)	5500±44	1700±20	1200±17
Cu ($\mu g\, g^{-1}$)	810±64	2100±140	1100±77
Cd ($\mu g\, g^{-1}$)	380±14	24±8	30±4
Cr ($\mu g\, g^{-1}$)	155±10	201±6	403±22
Ni ($\mu g\, g^{-1}$)	120±2	160±17	250±13

Data from Yoo, 1991.

Table 2 Composition (%) of stabilized WTE ash-concrete

Composition	*Mix BA*	*Mix BAFA*	*Mix BASR*
Bottom ash	66.6	35.0	43.4
Fly ash	–	23.2	–
Scrubber residue	–	–	18.6
Cement (type II)	16.8	18.4	19.5
Water	16.6	18.6	18.5
Lime ($Ca(OH)_2$)	–	4.8	–

(S/L) ratio (wt/vol) of 1:10, 1:100, and 1:1000. The actual weight/volume ratios were: 5g/50 ml for 1:10, 1g/100 ml for 1:100, 0.5g/500 ml for 1:1000. While these concentrations are equivalent to 1000mg/l to 100,000mg/l and generally far exceed normal suspended solids concentrations, they do represent a worst case situation and may in fact be representative of those concentrations found if lumps of ash accumulate in sediment.

The mixtures were then placed on a shaker to allow the reaction to occur at intervals of 0.5, 2, 8, 24, and 48 hrs. At the end of the leaching period, the aqueous phase from each reaction bottle was collected by filtering the mixture through a 0.45μm Millipore^R filter. The filtered solution was then analyzed for lead and cadmium using a Perkin-Elmer 5100 atomic absorption spectrophotometer (AAS) equipped with Zeeman background correction.

Three replicate samples from each leaching solution were analyzed. Matrix modifiers, i.e., NH_4NO_3, $(NH_4)_2HPO_4$, and Ni $(NO_3)_2$ were used in the analysis for cadmium and lead in sea water. National Institute of Standard and Technology (NIST) Standard Reference Material (SRM) 1633a fly ash and Multielement Solutions SRM 3171 and 3172 were used to determine the completeness of digestion of the ashes and to provide accuracy and quality assurance of the analytical methods.

By adding NH_4NO_3 to sea water, the sodium and chloride could be volatilized out of the graphite furnace at charring temperature near 500°C. Addition of NH_4NO_3 to sea water converts the sodium chloride (NaCl) into sodium nitrate ($NaNO_3$) and ammonium chloride (NH_4Cl) via the following reaction:

$$NaCl_{(aq)} + NH_4NO_{3(aq)} \rightarrow NaNO_{3(aq)} + NH_4Cl_{(aq)}$$

The sublimation temperature of NH_4Cl is 335°C, and NH_4NO_3 and $NaNO_3$ start to decompose at 210°C and 380°C, respectively, while the boiling temperature of NaCl is 1413°C.

In addition to matrix modifier, the use of the Zeeman device (applying a magnetic field to correct background absorption) eliminated most of the interferences normally encountered when direct sea water analysis is performed.

Sea Water Leaching Test on Stabilized Ash-Concretes

Metal leaching from the stabilized ash-concretes into surrounding sea water was examined following the method developed by Duedall *et al.* (1983). A solid cylinder of stabilized ash-concrete sample (5cm in diameter and 10cm in height; geometrical surface area 203cm^2) was suspended with monofilament line inside a polyethylene tank containing 2 litres of filtered sea water. Duplicate ash-concrete samples of each mix type were investigated. Each tank containing the ash-concrete sample was placed on a magnetic stirrer to generate a constant motion to the sea water. A 0.45μm membrane filter was placed over an opening in the cover of the tank to ensure aeration. The tank sea water was replaced with fresh sea water after the initial 3-day period, and then was replaced at two-week intervals for the remainder of the leaching period. The water samples were taken at the end of 1, 2, 3, 6, 9 and 12 day intervals in the initial 12-day period, then weekly during the next 6 weeks, and then sampled biweekly during the next 8 weeks. Collected water samples were filtered through a 0.45μm Millipore^R filter, acidified to pH 2 using Ultrex^R nitric acid, and stored for later analysis by AAS.

RESULTS AND DISCUSSION

Loose-Ash Leaching Study

Results of the loose ash-sea water leaching tests are expressed in terms of leachable fractions of lead and cadmium and are shown in Figures 1–5; leachable fraction, expressed as a percentage, is defined as the ratio of the mass of the metal leached to the total mass of the metal present in the original ash sample.

Loose fly ash

Release of lead from loose fly ash to sea water was rapid, occurring within 0.5 hr. Figure 1 shows that the leachable fraction of lead from fly ash decreased as S/L ratio increased. For S/L ratio of 1:1000, approximately 50% of total lead in the ash was released within 0.5hr of contact time. However, at S/L ratios of 1:100 and 1:10, less than 10% of leachable lead was released initially at the end of 0.5hr leaching period. However, for all the S/L ratios investigated, the apparent release of lead decreased with time.

It should be pointed out that the results showed that the leachable fraction decreased as the value of the solid/liquid ratio increased. This should not be construed to indicate that less metal in the dissolved form would be detected. On the contrary, higher concentrations of the solubilized element were measured in those solutions with higher concentrations of solids.

The observed rapid decrease in the release of lead from WTE fly ash at S/L = 1:1000 with increasing contact time may be due either to reprecipitation or to scavenging of lead by ash particles, or to the adsorption of lead on to the wall of the container used in the leaching experiment. Adsorption of dissolved lead on to the wall of the plastic bottle was ruled out, based on a control study involving addition of

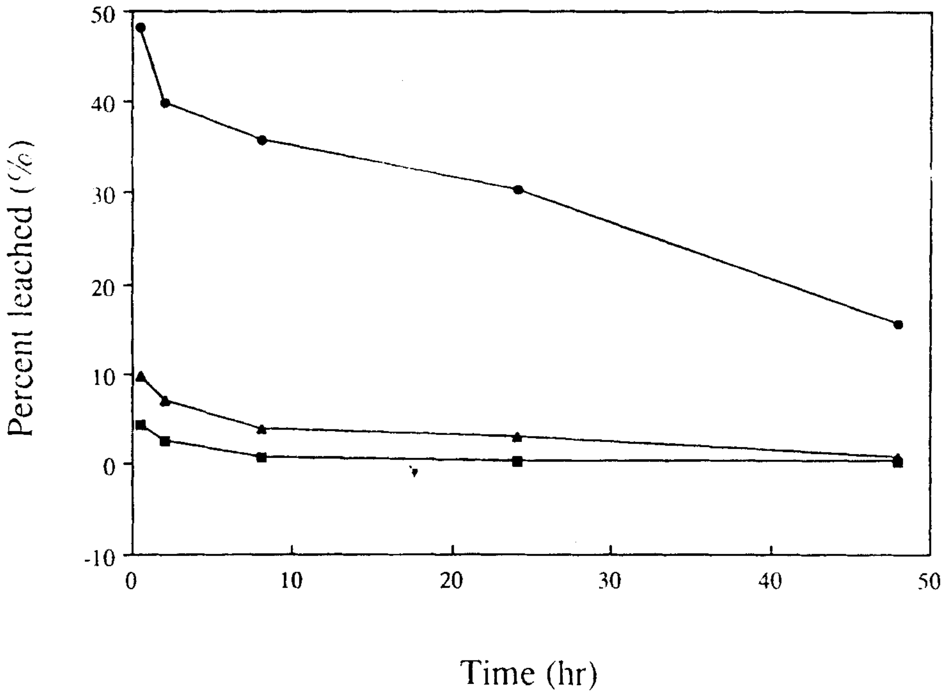

Figure 1 Leaching of Pb from loose WTE fly ash with time for S/L ratio of 1:1000 (●), 1:100 (▲), and 1:10 (■), respectively.

known amounts of lead to a solution which was treated in the same way as the test experiments. Thus, the large decrease in the amount of lead leached from the ash in the 48-hr ash-sea water leaching experiment was most likely due either to the scavenging of the dissolved lead by ash particles or to the precipitation of an insoluble lead compound, or to a combination of both.

Release of cadmium from loose WTE fly ash to sea water was also very rapid (Figure 2); greater than 95% of the leachable cadmium was released to sea water during the initial 0.5hr interval of experiment at S/L = 1:1000. Like lead, the amount of cadmium leached from the loose fly ash decreased with increasing S/L ratio. According to van der Sloot and Wijkstra (1990), cadmium in WTE ash is characterized by its strong solubility; that is, there is maximum leachable cadmium at the lower S/L ratio. For S/L ratios of 1:1000 and 1:100, cadmium was released from ash particles immediately (within 0.5hr). The cadmium remained in the solution as evidenced by the results from the 24 and 48 hr leaching tests. For S/L ratio of 1:10, the leachable fraction of cadmium from loose fly ash decreased with increasing contact time, indicating that relatively high concentration of ash particles in sea water likely leads to readsorption or scavenging of the released cadmium.

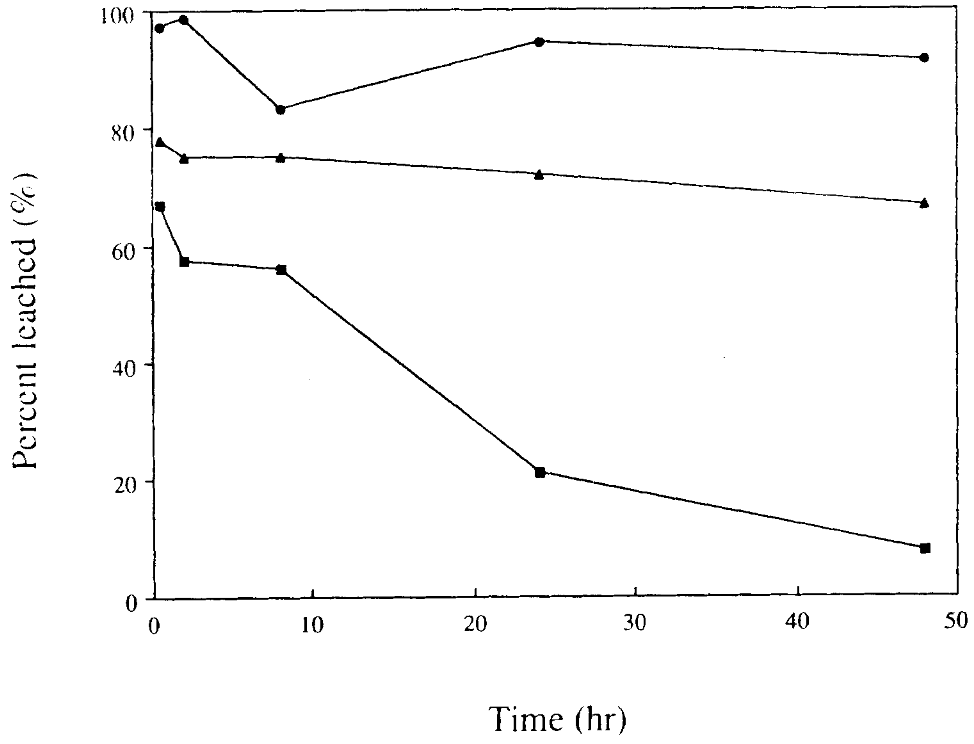

Figure 2 Leaching of Cd from loose WTE fly ash with time for S/L ratio of 1:1000 (●), 1:100 (▲), and 1:10 (■), respectively.

Loose bottom ash

Release of lead from loose WTE bottom ash is very minor compared to that from fly ash (Figure 3); less than 1% of leachable lead was detected in the sea water leachate containing ash particles at 1:1000 S/L ratio. No detectable lead was found in the leachate for both S/L ratios of 1:100 and 1:10.

Relatively low releases (< 1%) of cadmium from bottom ash was found for both S/L ratios of 1:100 and 1:10. However, larger releases (about 13–17%) of cadmium from loose WTE bottom ash occurred for S/L ratio of 1:1000 (Figure 4). Compared with fly ash, bottom ash is very much less reactive in terms of the release of cadmium to sea water.

Loose scrubber residue

Release of lead from loose WTE scrubber residue was not measurable for all S/L ratios investigated. However, release of cadmium from scrubber residue was observed for S/L ratio of 1:1000 (no detectable Cd was found in leachate for both 1:100 and 1:10 S/L ratios). Figure 5 shows that approximately 4.2% of leachable cadmium from scrubber residue was released within 0.5hr. Release of cadmium decreased to approximately 0.2% of that originally present after 2hr leaching. However, the release of cadmium increased to approximately 1.2% during the 48hr leaching experiment. The observed variation in the leachable fraction of cadmium may be due to changes in solution pH. Immediately after the addition of scrubber

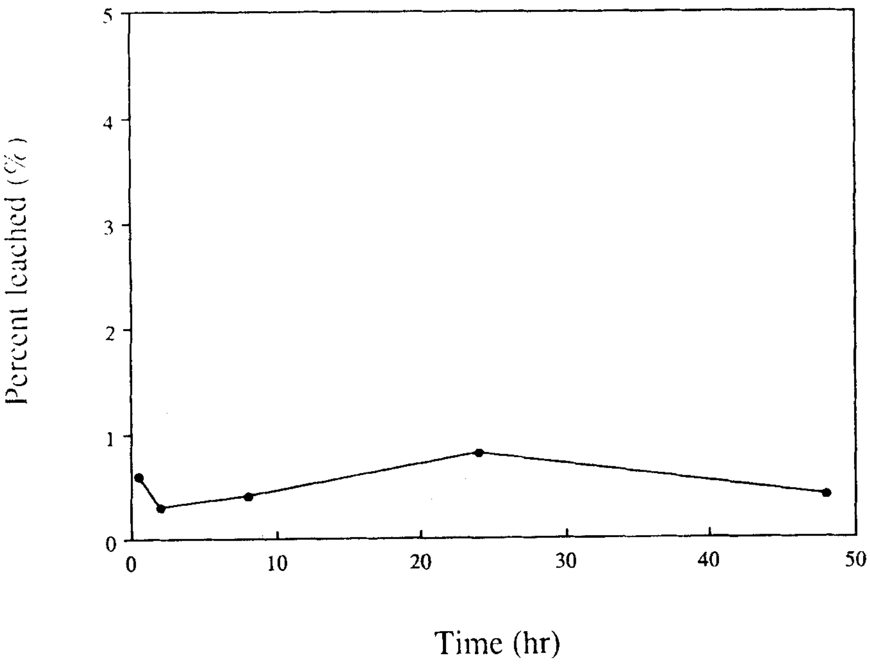

Figure 3 Leaching of Pb from loose WTE bottom ash with time for S/L ratio of 1:1000.

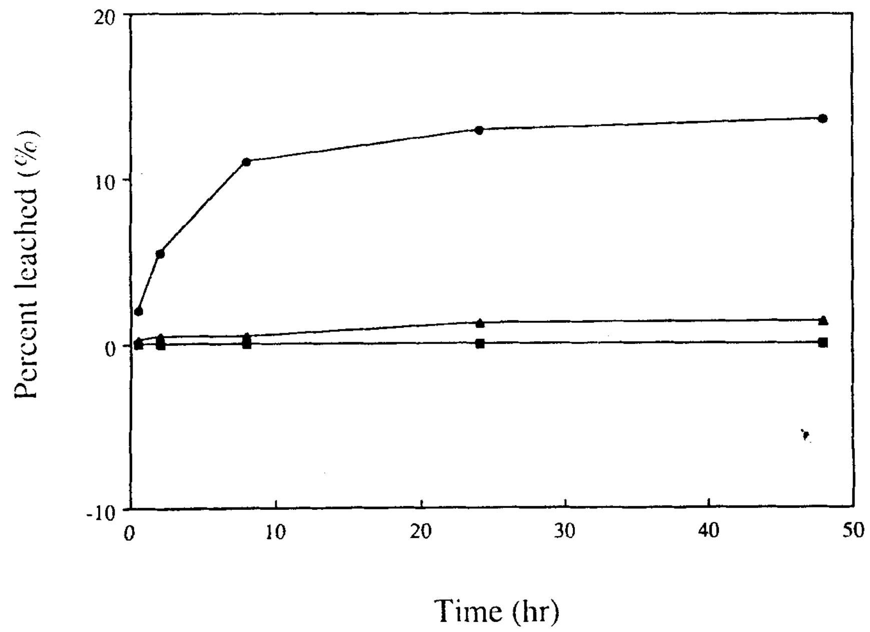

Figure 4 Leaching of Cd from loose WTE bottom ash with time for S/L ratio of 1:1000 (●), 1:100 (▲), and 1:10 (■), respectively.

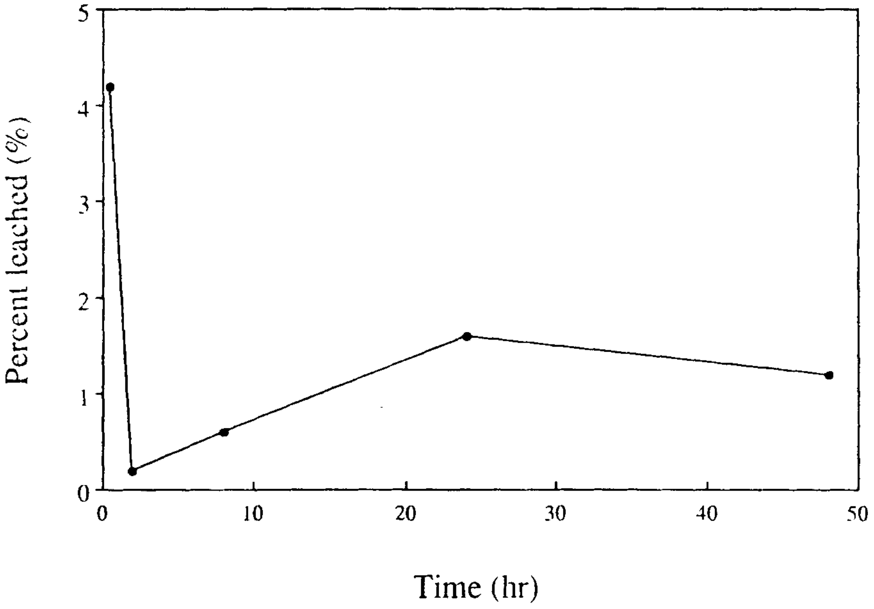

Figure 5 Leaching of Cd from loose WTE scrubber residue with time for S/L ratio of 1:1000.

residue to sea water, pH increased above 9 and then slowly returned to original sea water pH, i.e., pH = 8.1, after 48hr.

Overall, the loose WTE fly ash was found to be very reactive, while WTE bottom ash and scrubber residue were somewhat inert in sea water. The experimental results on loose ash also indicated that the alkaline or acidic characteristic of WTE ash and the pH of the solution might have some influence on the leaching behaviour of elements in the ash.

Stabilized ash-concrete leaching study

The results of stabilized WTE ash-concrete leaching study are presented here in terms of metal fluxes. While this paper reports primarily on lead and cadmium, the fluxes of calcium and magnesium from stabilized WTE ash-concrete to surrounding sea water were also investigated. Calcium was studied because it is responsible for the formation of compounds with cementitious properties in the stabilized ash products (Sheih *et al.*, 1990). Magnesium was examined because sea water magnesium is thought to replace a portion of the calcium in the stabilized ash-concrete (Edwards and Duedall, 1985) and also because of the formation of magnesium compounds inside the reacted stabilized ash-concrete has been reported and this process may have influenced the leaching of other metals (Hockley and van der Sloot, 1991).

The fluxes reported here represent average leaching (mmoles) of an element over a finite time period from the surface of the ash-concrete. The following equations were used for calculating metal fluxes:

Case I. change of sea water after sampling:

$$J_i = \{[C_t - C_o) \times V_t]/M\}/(A \times t)$$

Case II. no change of sea water after sampling:

$$J_{ii} = \{[C_t - C_o) \times V_t] - [C_{t'} - C_o) \times V_{t'}]\}/[M \times A \times (t - t')$$

where

J_i, J_{ii} = metal flux (mmoles mm^{-2} d^{-1})
C_t = metal concentration in tank solution at time t (mg l^{-1})
$C_{t'}$ = metal concentration in tank solution at previous sampling (mg l^{-1})
C_o = metal concentration in control sea water (mg l^{-1})
V_t = volume of sea water in the tank at time t (l)
$V_{t'}$ = volume of sea water in the tank at previous sampling (l)
M = molecular weight of metal (mg $mmole^{-1}$)
t = time since previous sea water replacement (days)
t' = time since previous sampling (days)
A = surface area of ash-concrete (mm^2)

The results of the study showed that, in general, release of Ca^{2+} from all types of mixes occurred only at the initial three days of the experiment (Figure 6). After three days, the WTE ash-concrete began to take up calcium from the tank sea water.

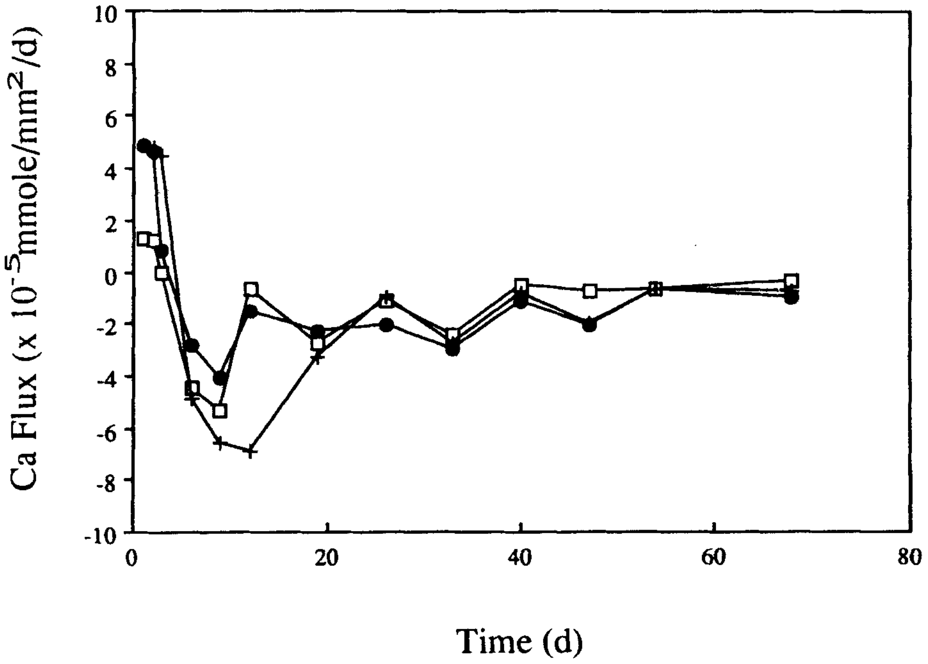

Figure 6 Flux of Ca in tank water containing mix BA (●), mix BAFA (□), and mix BASR (+).

Leaching of calcium at the earlier stage (< 3 days) in a tank leaching experiment may be attributed to the dissolution of soluble Ca-compounds, such as lime or gypsum, which are present on the surface of the stabilized ash-concrete and are "washed-off" from the surface due to solution agitation. Uptake of calcium from sea water (after 3 days) may be due to the precipitation of carbonate salts either on the surface of the products or within the pores of the block. When sea water penetrates into the block, the calcium present in normal sea water may react with aluminum-silicon components available in the block to form Ca-Al-Si compounds which could lead to improvement in the strength of sea water reacted ash-concrete. The formation of carbonate salts on the surface of stabilized ash blocks has been reported by Roethel *et al.* (1987) for another type of stabilized incineration ash, by Breslin (1986) for stabilized oil ash, and very recently by Hockley and van der Sloot (1991) for stabilized coal ash waste. The formation of ettringite, a Ca-Al-Si compound helpful in improving the strength of a stabilized ash waste, was observed in the coal ash-concrete and coal ash-sewage sludge concrete exposed to sea water (Roethel, 1981; Shieh and Roethel, 1989).

In contrast to calcium release during the initial three-day period, Mg^{2+} was removed from sea water; fluxes ranged from -12×10^{-5} mmole mm^{-2} d^{-1} to -7.5×10^{-5} mmole mm^{-2} d^{-1} within the first three days of the experiment. After three days no significant variation in magnesium flux was found.

Loss of magnesium, which systematically occurred for all types of stabilized ash-concrete during the initial three-day period, may be attributed to the formation of the mineral brucite ($Mg(OH)_2$) on the surface or within the pores of the block (Shieh *et al.*, 1990; Hockley and van der Sloot, 1991) or to the precipitation from the solution. The pore water pH in all stabilized WTE ash-concrete was relatively high (i.e., pH 12.0 for mix BASR; pH 12.4 for mix BAFA; pH 12.2 for mix BA) which exceeded the equilibrium pH (9.55) of brucite (Pytkowicz *et al.*, 1966). The tank water pHs were also greater than 9.55 during the initial three-day leaching period.

No detectable lead was found in any of the leachates containing different types of stabilized WTE ash-concretes. Cadmium in the tank leachate was either below or slightly above the detection limit ($0.1 \mu g\ l^{-1}$). It is clear that, when the lead and cadmium results are compared with the loose ash results, stabilized WTE ash-concrete is not reactive in sea water.

CONCLUSIONS

WTE residues are enriched in several trace metals of environmental concern. Fly ash was found to be the most reactive in sea water based on a comparison of leaching results for lead and cadmium. The ocean disposal of untreated WTE residues, especially fly ash, should not be considered. However, the WTE ash residues can be stabilized as a concrete form which is chemically stable in sea water; stabilization has significantly minimized leaching of cadmium and lead from WTE ash residues in sea water. Application of the stabilized ash-concrete in marine environment should be further investigated.

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