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Chemical Stabilization of Bottom Ash from Municipal Solid Waste Incineration and Prediction of DOC Leaching in Landfill Sites

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ABSTRACT
It is well known that it takes long time to stabilize landfill and meet the standard for leachate after landfilling is stopped. Moreover, the time required to meet the standard is not predicted. In this research, the rapid small-scale column test (RSSCT) was applied and was found to predict the dissolved organic carbon (DOC) leaching from municipal solid waste incineration bottom ash in a simulated landfill site. Acid washing with hydrochloric acid remarkably reduced the DOC leaching from bottom ash. According to the RSCCT, it was estimated that the acid washing would reduce the time for DOC stabilization of bottom ash by about 80% in a simulated landfill situation.

Keywords: acid washing, bottom ash, dissolved organic carbon, leaching behavior, RSSCT.

INTRODUCTION

The modern society is supported by the system of mass production, mass consumption, and mass disposal (Ministry of the Environment of Japan, 1999). In exchange for such prosperity, huge amounts of wastes have been discharged. In Japan, more than 75% of municipal solid waste (MSW) is incinerated and the residue is then deposited in landfills. Incineration of MSW is a management option, which has the potential to reduce the solid waste volume by 90%. Bottom ash from municipal solid waste incineration (MSWI) represents about 80% of the residues from incinerated MSW and contains a fraction of unburned organic matter and organic byproducts, which contain various hazardous organic substances (Johnson *et al.*, 1998, 1999) that may pose a threat to surface and groundwater quality (Nito and Ishizaki, 1997). Residual organic components which make up 1-2% of the MSWI material (Zhang *et al.*, 2004) also increase the biochemical oxygen demand (BOD) and chemical oxygen demand (COD) in the leachate from landfill sites through rainwater infiltration. In Japan, leachate from landfills has to satisfy the quality requirements defined by the "Waste Disposal and Public Cleaning Law", which establishes BOD and COD limits of 60 and 90 mg/L, respectively (Ministry of the Environment of Japan, 1999), for the discharge of leachate from landfill (leachate-controlled type). In addition to this limitation, many prefectures have more stringent effluent standards for landfill leachate as final storage quality (FSQ), and the representative standard value is 10 mg/L for both BOD and COD in Japan (Furuta, 2006).

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For the organic carbon sources, several studies (Brunner and Baccini, 1992; Brunner *et al*., 1987; Belevi *et al*., 1992) reported the importance of residual carbon in MSWI bottom ash. When 3,600 g of MSWI bottom ash was washed with water, the total organic carbon (TOC) concentration in the initial effluent was approximately 3,000 mg/L (Ishii *et al*., 2009). Ishii *et al*. (2009) also reported the importance of MSWI bottom ash as the source of dissolved organic carbon (DOC) in leachate for a long time and the effect of intensity, period and frequency of watering on the TOC elution from the bottom ash based on a numerical analysis. In addition, Tanaka *et al*. (2007) reported that solubilization of insoluble organics in bottom ash by biodegradation becomes the BOD source for a long time. Therefore, even after 18 years from the closing of landfill, more than 50 mg/L of BOD and COD has been detected in real landfill leachate from the landfill site where bottom ash has mainly been deposited (Fukui *et al*., 2006, Tanaka *et al*., 2007). Recently in Japan, the direct landfill of MSW is decreasing, and most of the MSW is deposited after incineration (Ministry of the Environment of Japan, 2008). The amount of bottom ash from MSWI was about 37 million tons in FY2006, and it was 77.6% of the total amount of MSW. Therefore, the importance of control of organic carbon, which gives origin to BOD and COD, is increasing. Tanaka *et al*. (2007) reported that the BOD/COD ratio in leachate was decreased to 0.1 after 19 years of ageing from around 1.0 at 5 years after closing the controlled-type landfill site in which the main content of landfilled waste was bottom ash. It indicated that biodegradable organics were degraded or leached in early stage. In addition, the residual non-biodegradable organic matter which could be detected as COD would be important as FSQ, because the standard value was the same for both BOD and COD as shown before.

To address the problem of DOC leaching from MSWI bottom ash, Misumi *et al*. (2006) established the wash-out waste landfill system (WOW system) to reduce the leaching potential of organic carbon from bottom ash by water washing. Mitsumi (2002) and Furuta (2006) evaluated the reduction of the period of water treatment to meet the BOD and COD limit (representative standard value: 10 mg/L) during a few years by using $1m^3$ of lysimeter, and reported that WOW system can reduce the period of water treatment from 35 to 20 years, including 15 years of landfilling (Furuta, 2006). We found that the organic carbon leaching from MSWI bottom ash was more remarkably reduced by acid washing than by water alone (Guimaraes *et al*., 2005). The acid washing could increase the percentage of reduction from 12% (by water washing) to 56%. This result indicates that the time required to FSQ is reduced by acid washing. In addition, acid washing can remove heavy metals because of the solubility for metals, therefore the elution potentials of toxic heavy metals in landfill would be also reduced by acid washing (Fedie *et al*., 2010). However, the time course of organic carbon concentration originated from acid-washed bottom ash has not been clarified yet. Therefore, it is necessary for its practical application to predict the organic carbon leaching behavior from MSWI bottom ash with and without pre-treatments with acid washing. However, the proper assessment and simulation by the lysimeter experiments having big volume, as Furuta (2006) conducted, includes time-consuming and expensive pilot-scale studies. Therefore, the prediction and evaluation by simulation was required.

The diffusion and elution could be estimated by using Fick's second law of diffusion

(Equation 1, where "*c*" is concentration, "D" is diffusion coefficient, "*t*" is time and "*x*" is position) (Guo T. *et al*., 2004).

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} \qquad (1)$$

Organic carbon leaching from MSWI bottom ash was found to be ruled by two mechanisms (Guimaraes *et al*., 2006). One is the leaching dependent on Ca leaching and the other is the leaching independent of Ca leaching. This indicates that the diffusion coefficient in Fick's law is not steady in a span of DOC elution from MSWI bottom ash and Fick's second law is not simply applied for the prediction of organic carbon leaching from MSWI bottom ash.

We focused on the rapid small-scale column test (RSSCT) developed for the prediction of the adsorption behavior of activated carbon by Crittenden *et al*. (1986). The fundamental concept of RSSCT was to scale the hydrodynamics and mass transport from full-sized media in a full-scale reactor down to smaller test media in a small-scale bench-top continuous flow test. It also predicts the performance of large columns from the result in RSSCT, not from physical properties and mathematical models like Fick's law, so extensive isotherm or kinetic studies are not required to obtain a full-scale performance prediction in RSSCT. Because the hydrodynamics and mass transport would be also parameters controlling the desorption of organic compounds from ash to continuous flow, we thought that RSSCT could be applied for the leaching of soluble materials from solids. The present work is an attempt to evaluate experimentally the possible application of RSSCT for predicting and simulating the organic carbon leaching behavior from MSWI bottom ash in a landfill site. Moreover, we evaluated the effectiveness of the acid washing treatment to shorten the time to reach stabilization through RSSCT.

MATERIALS AND METHODS

Bottom ash sample

Bottom ash was acquired from a MSWI facility in Higashi-Hiroshima City, Japan, in 2003. The plant incinerates 150 t/d of MSW and produces 7 t/d of bottom ash. This MSW mainly consisted of 53% paper, 17% plastic, 2% wood, 21% food waste and 7% incombustibles and other materials. The temperature in the combustion chamber was higher than 850ºC. About 20 kg of bottom ash was taken from the ash pit, homogenized and an aliquot of 2 kg was dried for 24 h at 100-105ºC which is the standard procedure for bottom ash treatment (Zhang *et al*, 2004). The final sample was stored in a glass bottle to be used for all experiments.

Table 1 shows the elemental composition of the bottom ash samples which was determined by acid digestion with nitric and hydrochloric acids (U.S.EPA, 1996). The resultant digestate was analyzed by inductively coupled argon plasma atomic emission spectrometry, ICAP-AES (model ICAP-575II, Nippon Jarrell-Ash) to determine the concentrations of Ca, Al, Fe and Si which were reported as the main components of bottom ash (Johnson *et al*., 1995). The relative abundance of each particle size fractions in the sample of bottom ash is shown in Fig. 1. The diameter mainly ranged from 1 to 15 mm.

Table 1 - Elemental composition and pH of MSW incinerator bottom ash samples.

	Ca	Fe	Al	Si
Contents (g/kg ash)	125	68	45	1

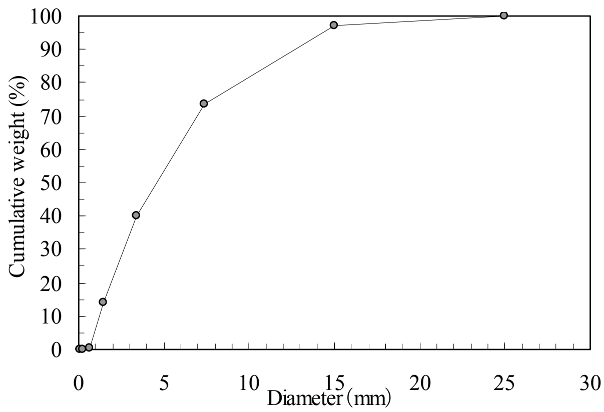

Fig. 1 - Relative abundance of particle size fractions in fresh bottom ash.

Washing method

Bottom ash was washed with hydrochloric acid (HCl) at pH 2 (Guimaraes *et al.*, 2005) to evaluate the capacity to decrease the organic carbon leaching and thus the time to reach stabilization in full-scale considerations. An automatic pH controller with acid addition (model TPD−51, TOKO Chemical Corporation) was used to keep the pH value at 2. Bottom ash sample of 20 g was placed in a glass beaker and acid solution of 400 ml was added at a liquid to solid ratio (L/S) of 20, and then the solution was continuously stirred at 20 ºC for 24 h. Up to 20 ml of acid solution was added to the system in order to keep the pH value at 2. Therefore, the L/S ratio was kept in the range 20-21. After 24 h, the solid fractions were separated from the solution by filtration through filter paper (Whatman 40). The solid was dried at 100-105^{o}C and subjected to column leaching experiments.

Column leaching experiments (RSSCT)

In order to evaluate experimentally the possible application of RSSCT in predicting the organic carbon leaching and thus, the time to achieve the chemical stabilization of bottom ash in a full scale, two different glass columns were selected to establish reliable scaling relationships. In the concept of RSSCT for granular activated carbon (GAC), the transport from the bulk liquid to the liquid film and the adsorption onto the GAC surface are acknowledged to be typically rapid and the overall rate of adsorption is usually controlled by either the liquid film transport or diffusion within the GAC particle. The leaching rate of DOC from bottom ash may also be approximated to be controlled by the liquid film diffusion around the ash particle, after dissolution or desorption from ash surface.

Scaling equations for RSSCT are developed by requiring that the dimensionless parameters of the small-scale and the full-scale adsorber be equal. By setting the dimensionless parameters of the RSSCT equal to those of a large-scale column, relationships between important design variables can be determined (Crittenden *et al.*, 1986). The surface diffusivity modulus, Ed, of the small-scale and large-scale columns are equated to match the spreading due to intraparticle mass-transfer resistance (Crittenden *et al*., 1986). The surface diffusion modulus (E_d) is a ratio of the diffusive mass transfer in the GAC to the rate of advection through the column. The equation that defines E_d is shown below.

$$E_d = \frac{DsDg\ \tau}{R^2} \quad (2)$$

where:
D_s = surface diffusion coefficient (cm^2/sec)
D_g = solute distribution parameter
τ = packed bed contact time (min)
R = GAC radius (cm)

Equating the E_d of the small-scale and large-scale columns a relationship is obtained between the EBCT values of the two columns, as shown below. This relationship incorporates the possible dependence of the intraparticle diffusivity on GAC particle size with the variable "*X*".

$$\frac{EBCT_{SC}}{EBCT_{LC}} = \left(\frac{R_{SC}}{R_{LC}}\right)^{2-X} = \frac{t_{SC}}{t_{LC}} \quad (3)$$

where:
$EBCT_{SC}$, $EBCT_{LC}$ = empty bed contact time of small and large columns (min)
R_{SC}, R_{LC} = GAC radius of small and large GAC (cm)
t_{SC}, t_{LC} = elapsed time to conduct the test for small and large columns (min)

Here, it is assumed that there is no dependence of intraparticle diffusivity on particle size, constant diffusivity "*X*" is 0 and equation 4 is obtained.

$$\frac{t_{SC}}{t_{LC}} = \left(\frac{R_{SC}}{R_{LC}}\right)^{2} \quad (4)$$

Analogous to RSSCT, the requirement in our study was that TOC desorption capacity of bottom ash could not depend on the bottom ash particle sizes used in the RSSCT and full scale. Fresh or washed bottom ash samples by HCl were placed in glass columns and leached by ultrapure water until the DOC concentration in the leachate presented negligible variation. Effect of intermittent water flow and inhomogeneous bed of landfill were ignored, and longer-term macroscopic change was evaluated here. In addition, the breakup of the matrix component of ash during long storage in landfill also causes inhomogeneous leaching of DOC. However, the breakup already occurred during washing operation. Therefore, the inhomogeneous leaching was also ignored in this evaluation.

Table 2 shows the specifications of column experiments and full-scale simulation as well as their operational parameters. Four hundred centimeters of landfill depth was assumed as a full-scale landfill site (60 m × 200 m), and 10 mm of particle diameter was selected as bottom ash size because the intermediate value of bottom ash diameter ranged from 0 to 20 mm (Fig. 1). In the simulation for full-scale landfill site by RSSCT, only rainy days were covered, that is, it was hypothesized that there was no flow of water in days free of rain. Therefore, the rainy days required to satisfy the FSQ of leachate were estimated from this RSSCT simulation. Weather data at

Higashi-Hiroshima City in 2006 was used for the simulation (Japan Meteorological Agency). The infiltration rate of full scale was calculated by the division of average annual amount of rainfall into the cross-sectional area with annual rainfall (1,839 mm) by the average number of days (20 d) with more than 30 mm of rainfall. On the basis of the hydrological parameters and landfill design, small (0.2-0.4 mm) ash particle was selected for small column experiments, and its EBCT was set based on equation 3. Then, the particle sizes in the range 0.8-1.0 mm were selected for the large column experiment, and its EBCT was also set from equation 3. All experiments were carried out at 20 ºC. Leachate samples were collected, filtered through 0.45 μm membrane filters (Millipore HA) and the clear filtrates were immediately analyzed for DOC by a TOC analyzer (TOC–5000, Shimadzu).

Table 2 - Specifications and operational parameters of column experiments and simulated full scale landfill site.

Parameters	Small column	Large column	Full scale
Length (Depth) (cm)	25	40	400
Cross-sectional area (cm^2)	1.76	20	1.2×10^8
Bottom ash filling (g)	15 (100%)	150 (100%)	- (100%)
Particle size of ash (mm)	0.2-0.4 (R_{SC})	0.8-1.0 (R_{LC})	10
Infiltration rate (L/d)	1.3	3.0	1.1×10^6
EBCT (h)	0.81	6.4	1044

RESULTS AND DISCUSSION

Prediction of Organic Carbon Leaching in Larger Scale – Validation of RSSCT

The column leaching tests for fresh ash to verify the scaling relationships are presented in Fig. 2. The design and operational parameters are listed in Table 2. Good correlation between DOC reduction profiles from the small and large columns was obtained. The difference of DOC concentration between small and large column was around 20 mg/L at the first few bed volumes, and less than 2 mg/L after that. The DOC concentration from each column was decreased to less than 10 mg/L around the bed volumes of 15 and leveled off at approximately 8 mg/L after the bed volumes of 25. The 15 bed volumes correspond to 12 h and 78 h in small and large column, respectively. The good correlation between DOC reduction profiles from the small and large columns guarantees the possible application of RSSCT to predict DOC leaching in a real landfill site.

Efficiency of Acid Washing – Simulation of Landfill Situation

The DOC leaching from fresh ash and washed ash by HCl was determined in small column experiments and is shown in Fig. 3. Acid washing was able to remarkably reduce the DOC leaching from bottom ash. Initial DOC concentration in the leachate from fresh and washed ash was around 350 mg/L (not plotted in Fig. 3) and 20 mg/L, respectively. DOC leaching from washed ash achieved stabilization in about 6 h of operation time and at very low concentration (<1 mg/L). Nevertheless, more than 12 h was necessary to equally achieve 10 mg/L in fresh ash. These results are corroborated by the findings of our previous research (Guimaraes *et al.*, 2005), which showed that HCl was the most efficient solvent for carbon extraction from MSWI bottom ash sampled at another plant and was able to reduce the DOC leaching potential by about

70%. Zhang *et al*. (2004) also found that HCl was able to extract the majority of organic carbon in bottom ash and hardly leached components, which comprised 40-80% of the organic matter in bottom ash.

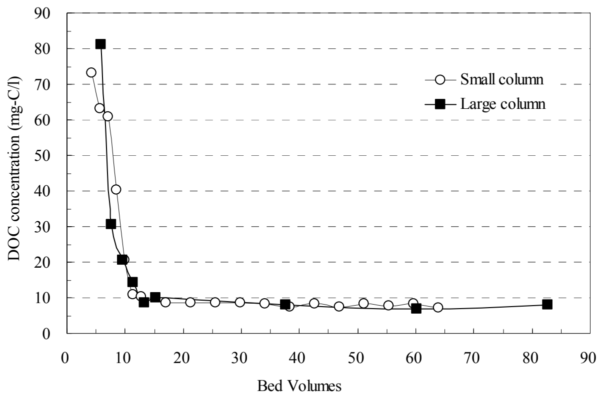

Fig. 2 - Comparison of the DOC elution profiles for the small and large column tests.

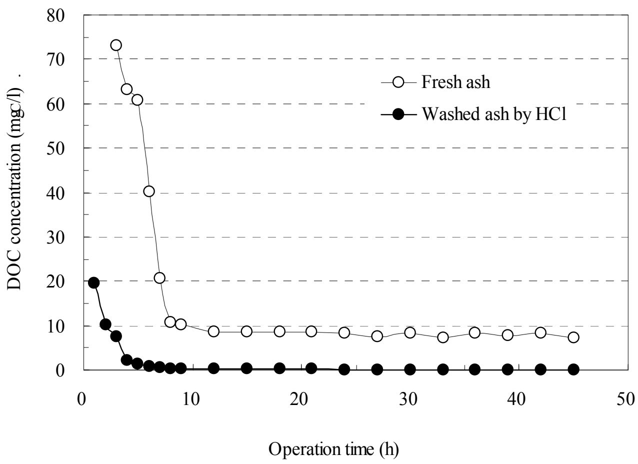

Fig. 3 - Efficiency of acid washing to decrease DOC leaching from MSWI bottom ash (result in small column experiments).

Fig. 4 shows the comparison between DOC leaching from fresh ash and washed ash by HCl acid solution as a function of operation time in the simulated landfill site (Table 2), predicted by the RSSCT (equation 4). The elapsed time was calculated assuming constant-rate leaching with 1,044 h of EBCT on the basis of 20 annual rainy days and 1,839 mm annual rainfall. Here, we focused on COD as FSQ in consideration of the biodegradation of organic matter during storage as mentioned in the introduction. Tanaka *et al*. (2007) reported that the COD/DOC ratio in leachate was around 1 at 5-19 years after closing of the controlled type landfill site in which the main content of landfilled waste was bottom ash. The representative standard value (FSQ) is 10 mg/L for COD in Japan (Furuta, 2006), therefore, we adopted 10 mg/L of DOC as a criterion in this evaluation.

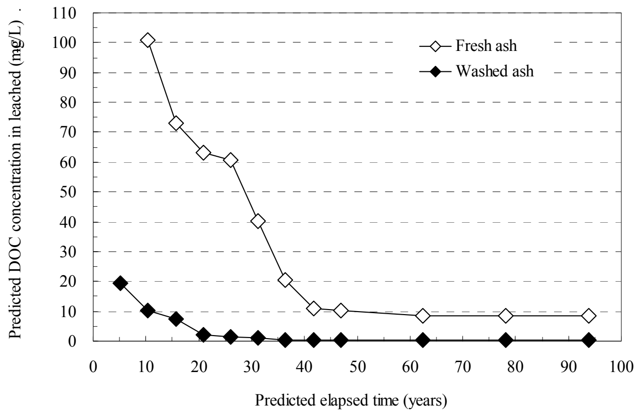

Fig. 4 - Predicted efficiency of acid washing in the simulated landfill site.

The leachate from acid washed bottom ash would satisfy with the FSQ, 10 mg/L at around 10 years, whereas it would require 50 years for fresh ash. This result indicates that the acid washing of bottom ash by HCl would reduce the time to reach chemical stabilization of landfill site and satisfy with the management level of DOC (10 mg/L) in the simulated landfill by about 80%. These results indicate that HCl washing presents high potential in practical applications aiming at reducing operational costs of landfill treatment facilities and their period of operation along with the landfill site-monitoring time (Guimaraes *et al.*, 2005).

CONCLUSIONS

The rapid small-scale column test (RSSCT) was applied and was found to predict the DOC leaching from MSWI bottom ash in a simulated landfill site. Acid washing with hydrochloric acid remarkably reduced the DOC leaching from bottom ash. According to the RSCCT, it was estimated that the acid washing would reduce the time for DOC stabilization of bottom ash by about 80% in a simulated landfill situation.

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