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Effect of Organic Matter on Leaching Behavior of Heavy Metals from Municipal Solid Waste Incineration Bottom Ash

by

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Abstract

Incubation experiments were carried out to study the leaching behavior of heavy metals (Cr, Cu, Pb and Zn) from municipal solid waste incineration (MSWI) bottom ash, as well as the effects of dissolved organic matter and pH on their leaching behavior. In order to enhance the effect of the organic matter, a mixture of MSWI bottom ash with crushed incombustibles rich in organic matter was also investigated. Two series of incubation experiments were performed under an aerobic condition for 1 year, and the temperatures were set at 37 °C and 50 °C, respectively. Leaching concentrations of dissolved organic carbon (DOC), selected heavy metals and major ions (Na, K, Ca, SO₄ and Cl) from MSWI bottom ash were investigated using Japanese Leaching Test No. 46 (JLT 46). The experiment results showed that dissolved organic matter from MSWI bottom ash was mainly composed of volatile fatty acids (VFA) and humic substances (HSs). The concentration of acetic acid was relatively steady in the early incubation stage, which contributed about 70 % of the total VFA, and was reduced suddenly when pH approached 9. Variation of HSs concentration in the leachate followed the same pattern with DOC, which at first showed an increasing tendency, subsequently decreased after reaching a maximum at the end of 24 weeks incubation period. The pH values of MSWI bottom ash decreased from its original value 12.2 to approximately pH 7-8 after a 1 year incubation process. Leaching concentrations of Pb and Cu from MSWI bottom ash decreased with the incubation time, especially in the first 4 weeks of the incubation period. However, the early incubation process promoted the leachabilities of Cr and Zn. At the end of the incubation process, the leaching concentrations of Cr, Zn and Cu decreased below 0.01 mg/L, while Pb still exceeded the legal limit of Japanese soil pollution. According to the analysis, leaching behavior of Cr, Cu, Zn and Pb had a strong correlation to pH value. And DOC partially contributed to the leaching of Cr and Cu. However, no relationship could be found between DOC and Pb or Zn.

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Keywords: MSWI bottom ash, Heavy metals, Leachability, Dissolved organic matter, Humic substances, Volatile fatty acids, Incubation

1. Introduction

Incineration is a preferable technology for municipal solid waste treatment in Japan with limited landfill capacity. In the past ten years, about 78 % of the total weight of municipal solid waste (MSW) has been incinerated yearly in Japan¹⁾. Hence the main composition of landfill waste is municipal solid waste incineration (MSWI) residues, mainly bottom ash. Organic matter in MSW can not be completely extinguished during the incineration treatment process, and around 0.2-5 % of unburned organic carbon remains in the MSWI residues²⁾. In addition, the landfill sites are usually composed of around 10 % crushed incombustibles (including shreds of waste electrical equipment, glass, ceramics, and unburned furniture, etc.) in Japan. Actually, most of the organic matter in the landfill sites is in the incombustibles, although it does not account much of the landfill waste. Some of organic matter in MSWI residues will be biodegraded and/or transformed to refractory organic matter such as humic substances (HSs) over time in the landfill³⁾. It is well known that HSs have a great influence on the fate of heavy metals in water and solid matrix because of their large adsorption capacity^{4, 5)}.

Previous studies have reported that when MSWI bottom ash experienced ageing or carbonation process, the leachabilities of heavy metals decreased dramatically⁶⁻⁸⁾. Carbonation reaction caused by the dissolving CO₂ from outside contributes to the heavy metals' immobilization as it decreases pH from alkaline to moderate level. In addition, acidities from the biological degradation process should be considered in the leachabilities of heavy metals, which are of the same order of magnitude as the acid neutralization capacity of bottom ash^{9, 10)}. Considering the large adsorption capacity of HSs for heavy metals, it is generally thought that HSs produced biologically from residual carbon in MSWI bottom ash might also have an effect on heavy metals leaching behavior.

This study was focused on the leaching behavior of Cr, Zn, Pb and Cu during the incubation process. Then two main factors that had effect on the leachabilities of heavy metals, pH and DOC were emphasized. The composition of the dissolved organic matter was considered to be HSs and volatile fatty acids (VFA). Also the potential effect of added crushed incombustibles, which were richer in organic matter than bottom ash, on the leaching behavior of heavy metals, and other characteristics of MSWI bottom ash were also investigated.

2. Materials and Methods

2.1 Materials and sampling

Fresh quenched MSWI bottom ash was taken from a solid waste incineration plant in Japan. Crushed incombustibles were obtained from a recycle center in Japan. The MSWI bottom ash and crushed incombustibles were sieved to 4.75 mm and metals particles were removed from them before bringing back to the laboratory.

2.2 Incubation experiment

Three kinds of samples were prepared for the incubation experiments: bottom ash (BA), crushed incombustibles (CI), and mixture of 80 % bottom ash with 20 % crushed incombustibles in weight (MIX). The above samples were filled in stainless steel containers with untight covers to hinder the water evaporation and to ensure ventilation. The containers were put in constant

temperature ovens. The materials and incubator apparatus are shown in **Fig. 1**. The incubation experiments were continued for 1 year under aerobic conditions. Every sample was incubated under two constant temperatures: 37 °C and 50 °C, respectively. During the incubation process, the water content of the samples was adjusted at 30 % by adding distilled water every two or three days, simultaneously mixing the samples. However, it was found that the samples incubated under 50 °C were dried more quickly than that incubated under 37 °C due to the increased water evaporation rate at higher temperature. Sub-samples were taken out from the containers after 2, 4, 8, 12, 18, 24, 32, 36, 44 and 52 weeks of incubation period. Immediately after sampling, these samples were air-dried, later grinded and sieved through a 2 mm mesh. The pretreatment samples were kept in closed plastic bags and saved under 4 °C till the time of analysis.

2.3 Characterization

Loss of ignition (LOI) of the sample dried at 105 °C was determined by combustion at 600 °C for 3 hours in an oven. The selected element concentrations in the MSWI bottom ash were determined by inductively coupled plasma spectroscopy (ICPS-7000, Shimadzu Co.).

Leaching characteristics of the samples were investigated using Japanese Leaching Test No. 46 (JLT 46). Samples were conducted in a liquid to solid ratio of 10 L/kg, stirring at 200 rpm for 6 hours, and finally the supernatant liquids were filtrated with 0.45 µm membrane filters in according with JLT 46. The pH, EC and ORP values of the leachate were determined with pH meter (F-23, HORIBA), EC meter (DS-14, HORIBA), and ORP meter (F-51, HORIBA), respectively. The concentration of water dissolved organic carbon (DOC) was determined by total carbon analyzer (TOC-V, Shimadzu Co.). The major ions (Na, K, Ca, SO₄ and Cl) were quantified by ion chromatography (DX-120, Dionex Co). The concentrations of Cr, Cd, Ni, Zn, Cr, Cd, Pb, Fe, and Mg in JLT 46 leachate were analyzed by inductively coupled plasma spectroscopy.

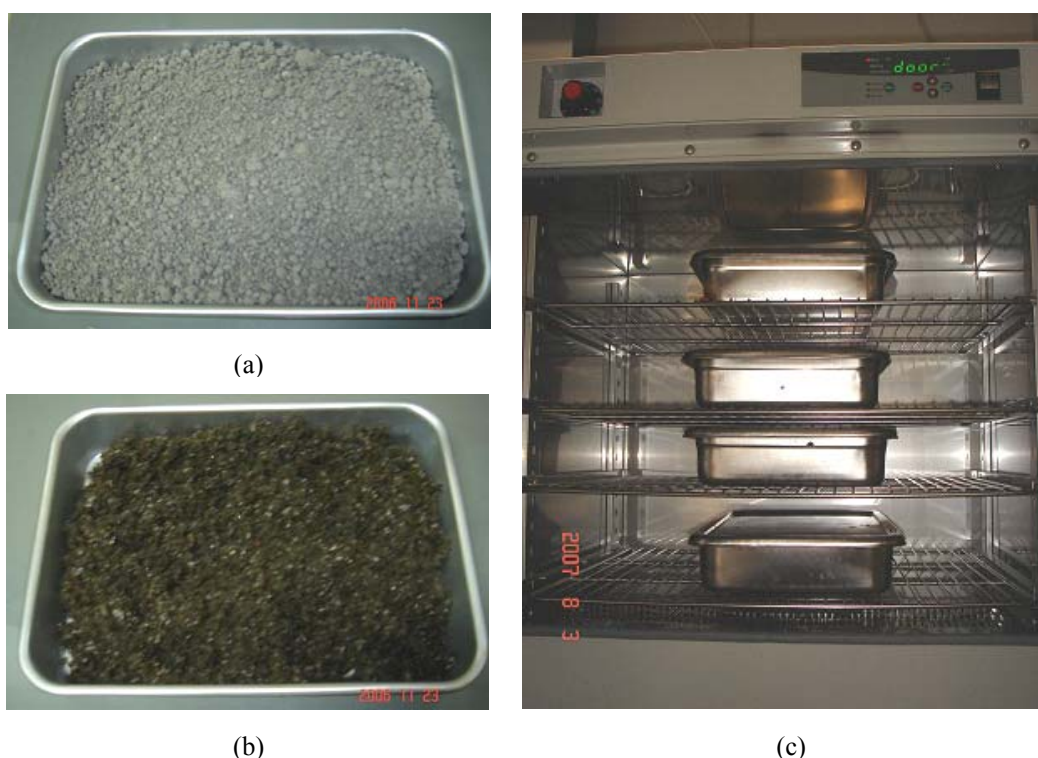


Fig.1 Materials and apparatus applied in this incubation experiment. (a) bottom ash; (b) crushed incombustibles; (c) containers and incubation apparatus.

HSs were extracted from JLT 46 leachate of fresh and incubated MSWI bottom ash, according to the procedure recommended by International Humic Substances Society (IHSS). The leachate was acidified to pH < 2 by 6 M HCl, then it passed through a column filled with DAX-8 resin (Supeline TM, Supelco Co.). HSs were eluted from the DAX-8 resin by 0.1 M NaOH, and their concentration in carbon was determined by a total carbon analyzer (TOC-V, Shimadzu Co.).

Volatile fatty acids (including acetic acid, propionic acid, i-butyric acid, n-butyric acid, i-valeric acid, n-valeric acid, i-caproic acid and n-caproic acid) in the leachate were measured by gas chromatograph (GC-2014, Shimadzu Co.). The analysis were performed with a TIC Column: length 30 m; flow rate: 40 cm/sec; carrier gas: helium; oven temperature: 45 °C to 225 °C; temperature increase rate: 10 °C/min; and injector/detector temperature: 250 °C.

3. Results

3.1 Characters of the bottom ash

Table 1 shows the concentrations of selected elements and organic matter in the fresh MSWI bottom ash and leaching concentration in the JLT 46 leachate of the MSWI bottom ash, as well as the legal limit of heavy metals regulated by Ministry of the Environment, government of Japan. The concentrations Cu and Pb in the bottom ash, and the leaching concentration of Pb were higher than the legal limit. Although there is no legal limit for Zn, Cu and Ni leaching, usually it is thought that the leaching concentration below 0.01 mg/L is acceptable. So the fresh MSWI bottom ash has potential risk to the surrounding environment in the process of reuse or landfill.

LOI of bottom ash was 5.1 %, however only 25 % of LOI was caused by organic matter¹⁾, so the organic matter content was estimated to 1.28 % (i.e. 12,800 mg/kg). LOI of CI was 3.6 %, which could be assumed as its real organic matter content. Organic matter content in CI was approximately 3 times to the organic matter in BA.

Table 1 Concentration of selected elements and organic matter in MSWI bottom ash and JLT 46 leachate, together with legal limit of heavy metals in for surveying soil pollution.

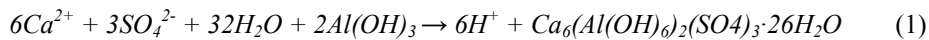
Metals	Concentration in MSWI bottom ash (mg/kg)	Legal limit* (mg/kg)	Concentration in JLT 46 leachate (mg/L)	Legal limit* (mg/L)
Zn	1,490	-	<0.01	-
Cu	1,150	<125**	0.39	-
Pb	295	<150	0.04	<0.01
Cr	84	<250	0.01	<0.05
Ni	21	-	<0.01	-
Cd	4	<150	<0.01	<0.01
Ca	142,000		437	
Al	54,700		94.8	
Fe	29,200		0.03	
Na	16,400		560	
Mg	9,290		<0.01	
Mn	261		<0.01	
Organic matter	12,800		18.3	

* Environmental Quality Standards for Soil Pollution Regulated by Ministry of the Environment, Government of Japan

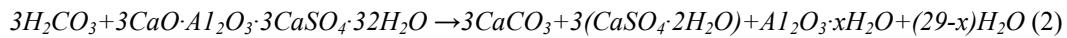
** In soil for agricultural land (paddy fields only)

3.2 pH, Eh and EC

The landfill site containing MSWI bottom ash disposal generally has a high pH and alkalinity in the beginning. Usually, pH drops down to neutral after several years by absorption carbon dioxide from atmosphere or produced from inner organic matter degradation, and permeation of acid rain precipitation. The process was simulated by the incubation experiment. As shown in **Fig. 2**, the pH values of BA and MIX decreased continuously during the incubation period. The pH decrease process was described as 3 phases according to the pH range: phase I (initial value - pH 11), phase II (pH 11 - 8.5) and phase III (pH < 8.5). And in each phase, the pH drop was controlled by different mechanism. During phase I, pH decreased due to the formation of ettringite and the mechanism is illustrated in reaction (1)¹². Simultaneously, Ca and SO₄ concentrations in the leachate reduced (see **Fig. 8**). It took 18-24 weeks to complete this phase for BA and MIX in this incubation experiment.



Subsequently, the pH decrease was mainly caused by the carbonation reaction with CO₂. In phase II, ettringite was dissolved and transformed into calcite and gypsum as described in reaction (2). Correspondingly the concentrations of Ca and SO₄ also showed and increase (see **Fig. 8**). The released SO₄ contributed to the pH decrease. Until bottom ash approached equilibrium with respect to CO₂ and calcite, the pH value reached to 8.0~8.5¹³. In this phase, the pH drop rate was much different at the two temperatures. It has been mentioned that the water content of the sample incubated under 50 °C decreased more quickly than that under 37 °C. As we know, carbonation reaction requires water. So the pH of BA sample incubated under 50 °C decreased slower comparing to that of temperature 37 °C, due to the absent of water.



In the phase III, pH entered a moderate condition, which enhanced the microbial activity. The pH decrease caused assumably by the speedy biodegradation of organic matter.

The initial pH of CI was 9.2, and it increased to 10 during 2 weeks of incubation period, followed by a decrease for the samples incubated under both temperatures. From the 12th week, pH of sample incubated under 50 °C fluctuated in the range of 9.4~10, while pH of sample under 37 °C varied in the range of 8.2~9.2. MIX had the same pH character with BA, and the difference was that the pH decrease rates were same under both temperatures. Considering that the MIX was a combination of BA and CI, this result might was due to added CI.

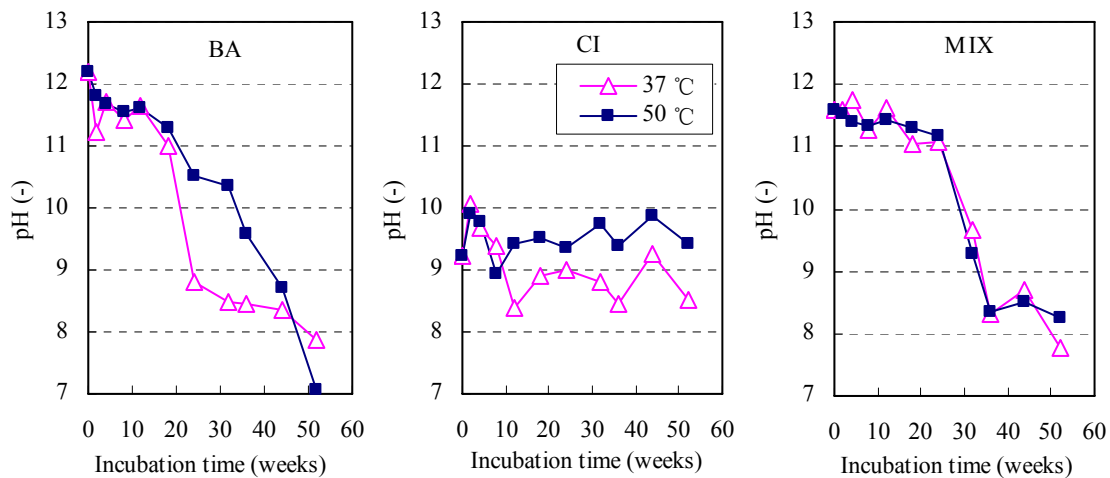


Fig. 2 pH of JLT 46 leachates with incubation time.

Eh value was derived using determined ORP, and it was positive in each sample which indicated that the entire incubation process underwent in an aeration condition (**Fig. 3**). Initial Eh was in the range of 200-300 mV, then increased to a range of 300-400 mV and remained steady until the 44th week, which was favorable for aerobic growth. Eh of all the samples showed a dramatic decrease from the 44th week to the 52nd week, during which pH values of BA and MIX decreased to around 8. When pH reached 8, metabolism of microorganism became active, with fast oxygen consumption and accumulation of reduction matrix, which caused the fall of the Eh value. Under both incubation temperatures, the Eh values were quite consistent.

EC values of BA and MIX were in the range of 3-5 $\mu\text{s}/\text{cm}$ (**Fig. 4**), which was related with the concentrations of major anions (Cl and SO_4) and cations (Ca and Na) in the leachate from the samples. EC promptly decreased in the early incubation stage, caused by the decrease of Ca and SO_4 , followed with a short constant period. In the later incubation period, EC value was entirely controlled by the Cl concentration (Cl data was not shown in this paper). In the case of Cl sample, EC was in the range of 0.3-1.0 $\mu\text{s}/\text{cm}$ and increased gradually with incubation time. And increase rate was slightly higher under 50 °C than that under 37 °C.

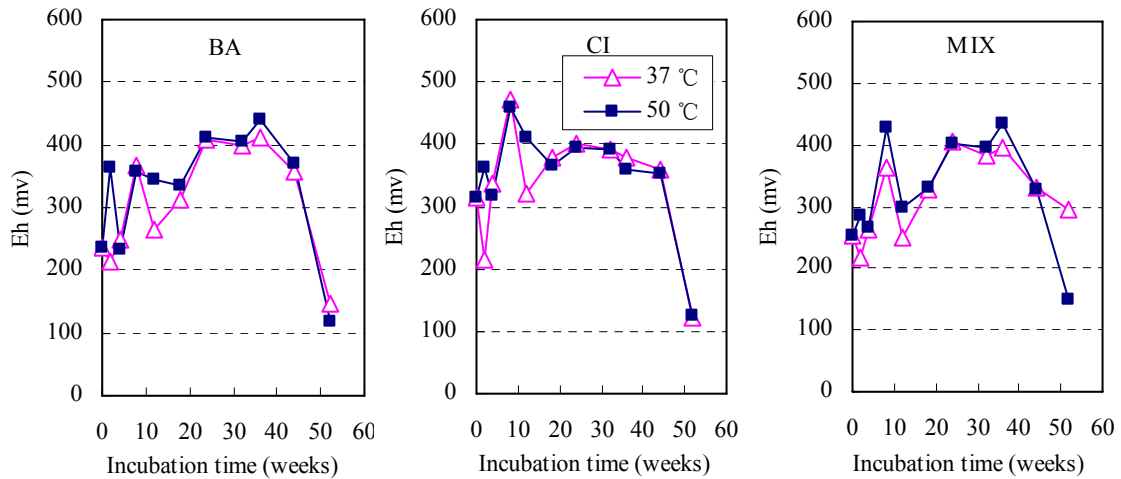


Fig. 3 Eh of JLT46 leachate with incubation time.

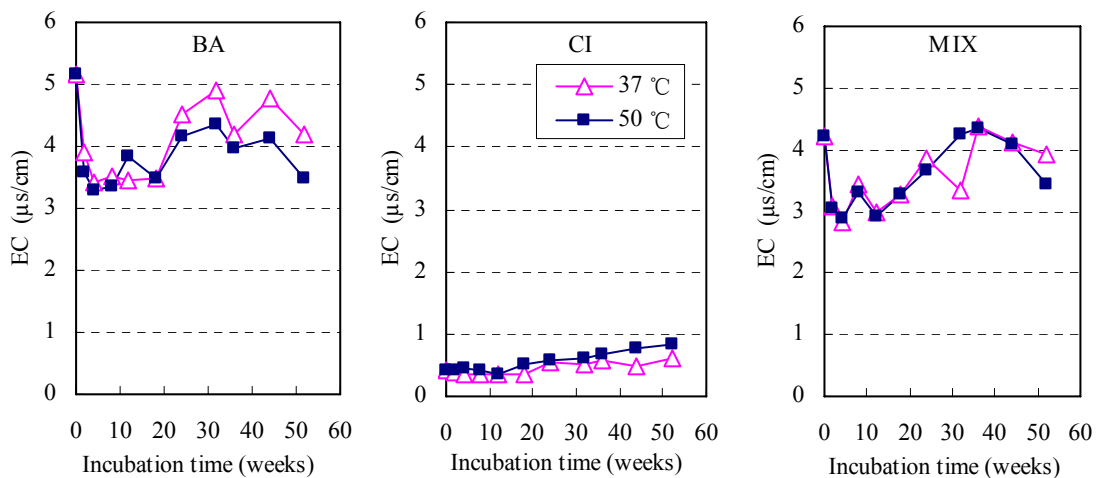


Fig. 4 EC of JLT46 leachate with incubation time.

3.3 Evolution of dissolvable organic matter

The DOC concentrations of the samples were shown in **Fig. 5**. The DOC concentrations of BA incubated under both temperatures kept constant in the early stage, and there was a discrepancy at the end of 8 weeks incubation period. The DOC of BA under 50 °C began to increase and reached maximum peak at the 24th week, then it decreased continuously till the end of the incubation process. DOC concentration of BA under 37 °C showed a decreasing trend, although there was a shoulder peak at the 24th week. DOC of MIX was relatively constant under 37 °C, while there was a large quantity of dissolved organic carbon generated since the end of 18 weeks incubation period. One of the components of CI contained microorganism, which made it possible for MIX to generate DOC quickly (**Fig. 5**). MIX had a high value DOC comparing with the BA due to the effect of CI. Generally, DOC concentrations in the three kinds of materials were higher at 50 °C comparing to that at 37 °C after the several weeks of incubation.

The HSs concentrations in the JLT 46 leachate of BA were shown in **Fig. 6**, as well as DOC concentrations. It was obvious that HSs followed the same evolution pattern with DOC. HSs were one of the major fractions in DOC, which accounted for about 30-60 %. At the end of the incubation process, HSs mostly contributed to DOC concentration.

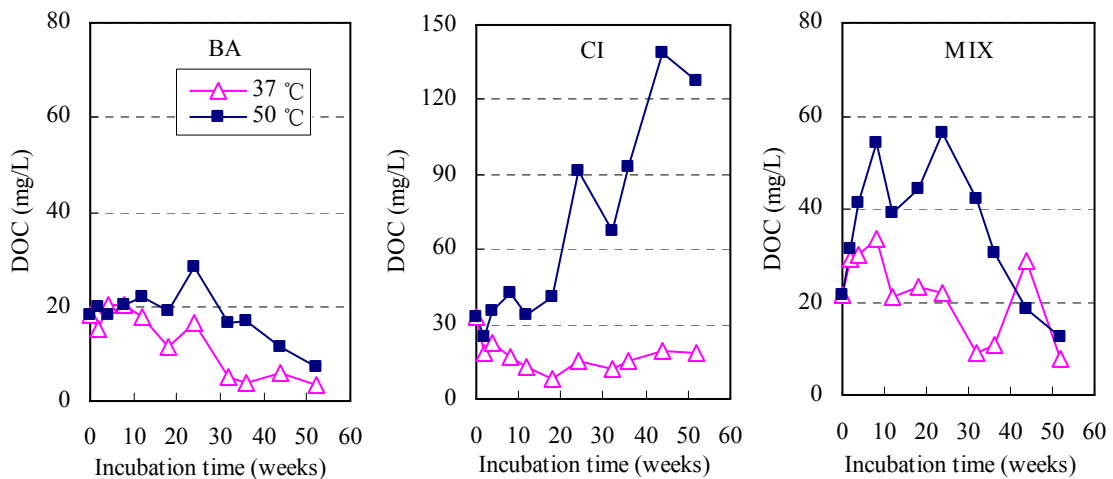


Fig. 5 DOC concentration in JLT46 leachate with incubation time.

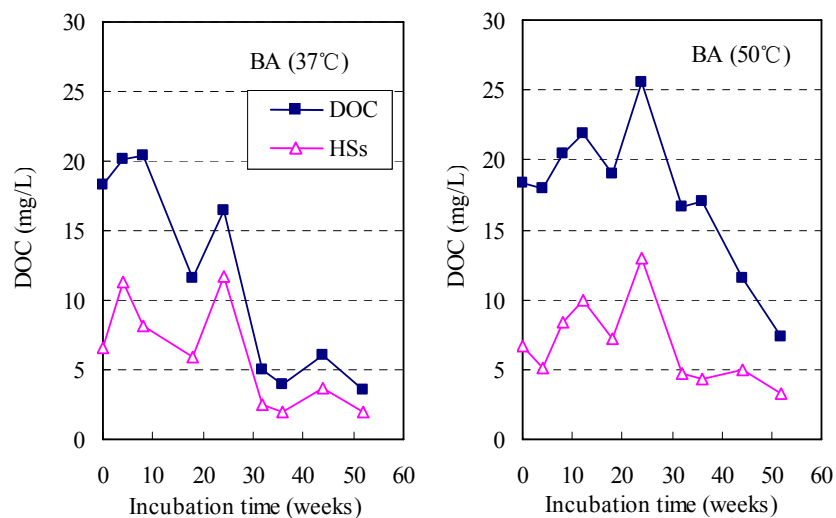


Fig. 6 DOC and HSs concentration in JLT46 leachate with incubation time.

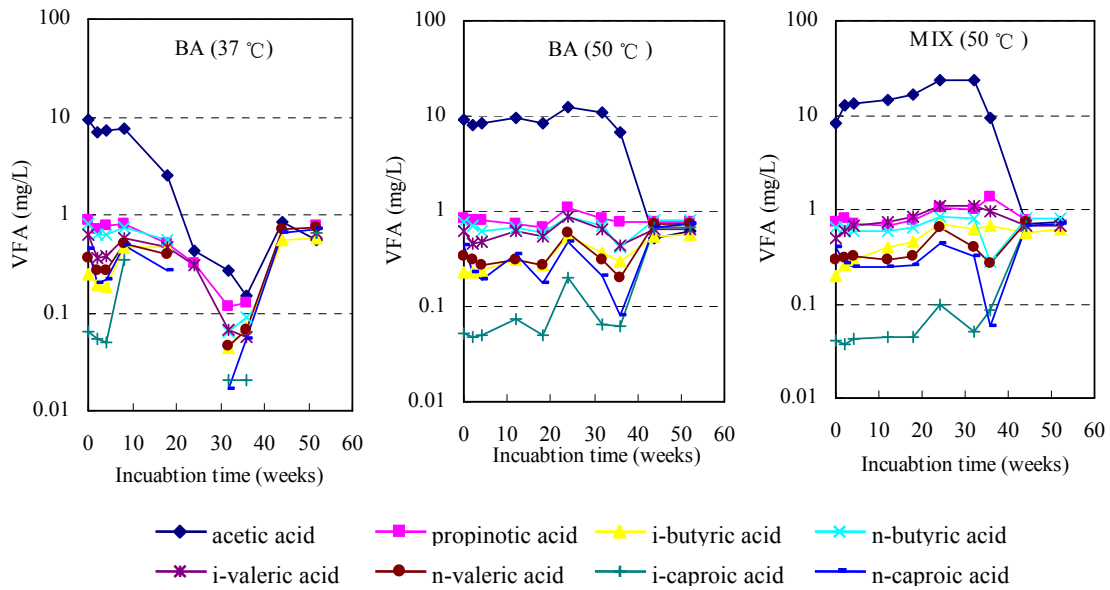


Fig. 7 Volatile fatty acids concentration in JLT46 leachate with incubation time.

Volatile fatty acids (VFA) were index of organic matter degradation. It was found that VFA were one of the main components of dissoluble organic matter in leachate besides HSs. And acetic acid was the major VFA, followed by propionic acid, i-butyric acid, n-Butyric acid, i-valeric acid, n-valeric acid, i-caproic acid and n-caproic acid, as shown in **Fig.7**. Acetic acid concentration reduced greatly when pH reached 9 (refer **Fig. 2**), and at the same time the concentrations of the most other kinds fatty acids slightly increased.

3.4 Leaching concentrations of major ions and heavy metals with the incubation time

3.4.1 Ca and SO₄

Ca, Na and K were the major cations in the JLT 46 leachate and SO₄ and Cl were the major anions. Na had a concentration range of 400-900 mg/L, and K was in the range of 60-300 mg/L. The concentration of Cl gradually increased from 700 mg/L to 1400 mg/L during the incubation process. Here, we only show the Ca and SO₄ values (**Fig. 8**), because they have close correlation with the pH value. Fresh bottom ash had a Ca concentration of about 400 mg/L, which was decided by the solubility of Ca(OH)₂. However, Ca concentration in BA and MIX decreased quickly during the first 12 weeks of incubation period due to the formation of calcite and ettringite, then the concentrations increase of Ca and SO₄ due to the dissolve of ettringite. Leaching concentration of Ca well correlated with SO₄ in the incubated BA and MIX samples. For fresh bottom ash, SO₄ existed in the phase CaSO₄ with a low solubility. So the initial concentration of SO₄ in the leachate was low. However it increased rapidly after start the incubation.

The concentration Ca in the JLT 46 leachate of CI was relatively stable, exception of a decrease in the initial stage (**Fig. 8**). The leaching concentration of SO₄ in CI increased gradually with the incubation time. Ca and SO₄ in the leachate of MIX showed the same trend with that of BA. Even the leaching behavior of BA and CI was so different, but CI had insignificant effect on the leaching concentration of Ca and SO₄ from MIX.

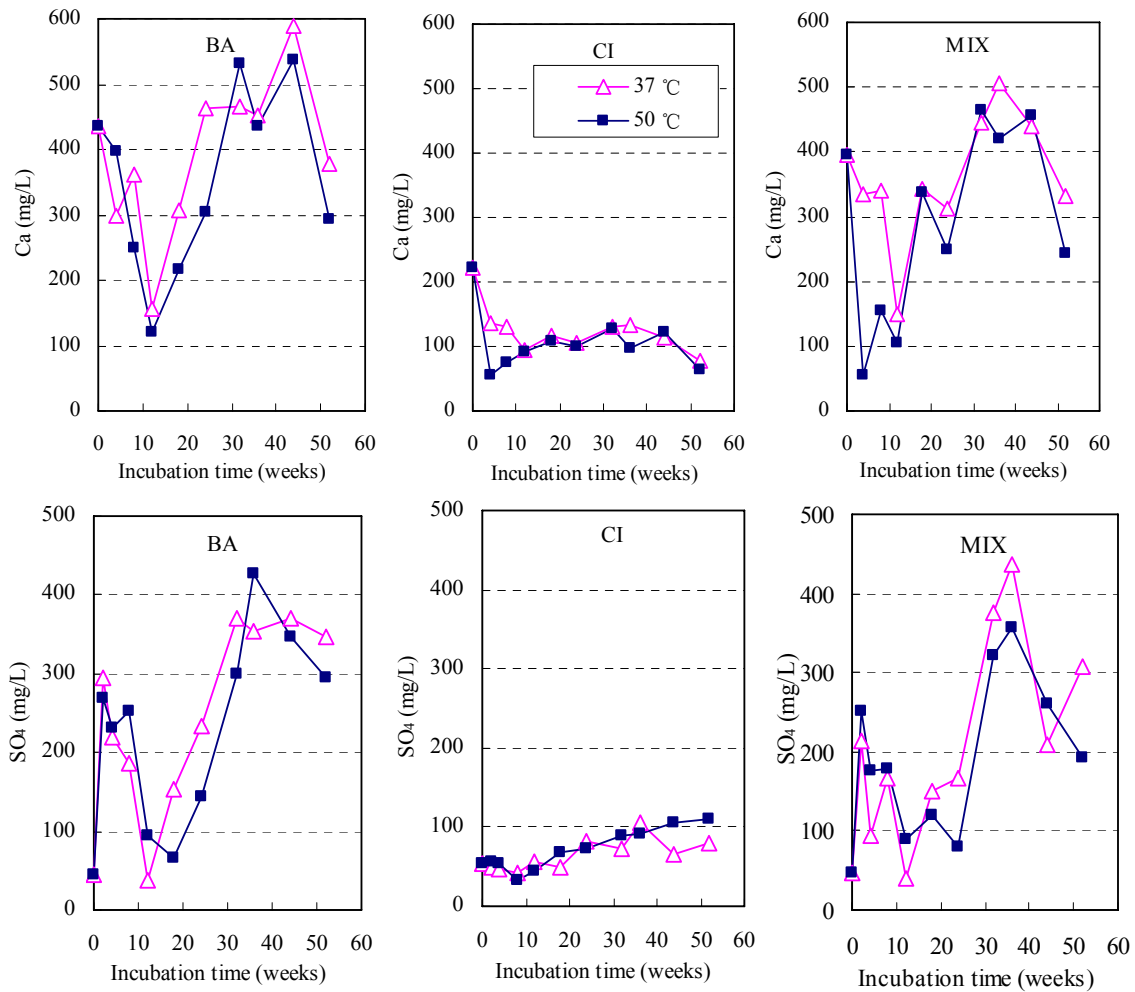


Fig. 8 Concentrations of Ca and SO_4 in JLT 46 leachate with incubation time.

3.4.2 Cr, Zn, Pb and Cu

The leaching concentrations of Cr, Pb, Zn and Cu from the samples were shown in Fig. 9. In the case of the leaching concentration of heavy metals lower than the detectable limit of 0.01 mg/L, it was set as 0.01 mg/L (see Fig. 9, Fig.11 and Fig.12). At the beginning, the leaching concentration of Cr from BA was approximately 0.01 mg/L. However, incubation process promoted their leachabilities obviously. Then, Cr concentration decreased to below the limit value after 32-36 weeks of incubation period. The leaching concentration Cr from CI was lower than the detectable limit during the whole incubation process. Leaching concentration of Cr from MIX followed the same evolution tendency as BA, but was lower than BA. That was caused by the dilution of added CI.

The leaching concentration of Pb from BA was quite high, and it decreased after incubation. On the contrary, the leaching concentration of Pb from fresh CI was low and it increased continuously during the incubation process. Because of the influence of added CI, Pb in the leachate of MIX was comparative higher than that of BA in the late incubation stage.

Similar to Cr, the initial leaching concentration Zn was lower than the limit, and there was a slight increase in the early incubation stage. Zn leaching concentration was quite high in CI that was incubated under 50 °C, especially from the 36th week to the 44th week, whereas it had no effect on the leaching concentration of Zn from MIX.

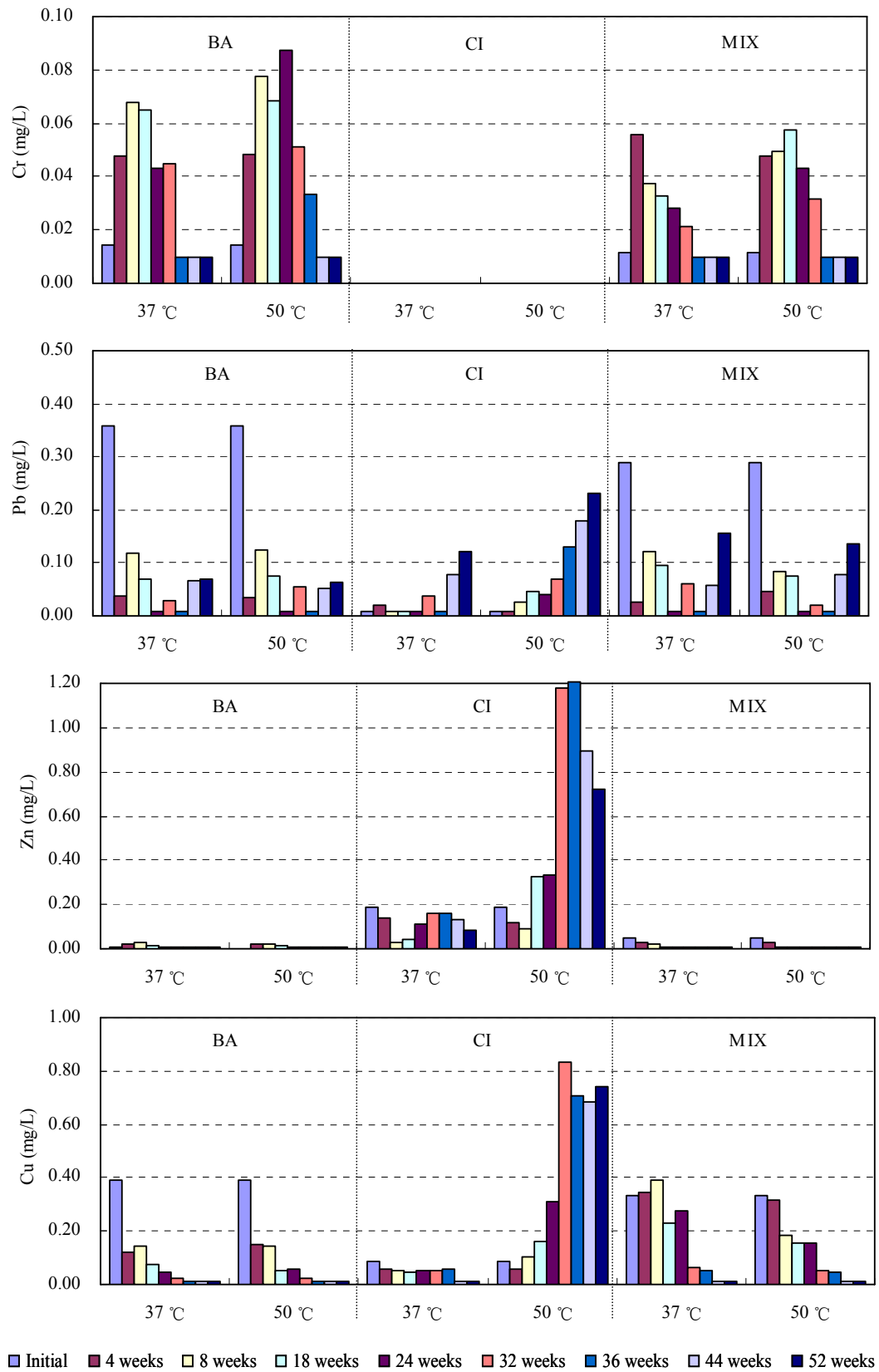


Fig.9 Concentrations of Cr, Zn, Pb and Cu in JLT 46 leachate with incubation time.

The leaching concentration of Cu from BA showed a decrease tendency continuously during entire incubation period. And Cu concentrations of CI were dissimilar when CI was incubated under the different, which Cu leached out more easily under the higher temperature. On the other hand, the leaching concentration of Cu from MIX was higher than that from BA in the term of the 4th week to 36th week.

Temperature had no effect of the leaching behavior of heavy metals in BA and MIX mentioned above. While higher incubation temperatures could promote the leachabilities of Zn, Pb and Cu from CI. Ni and Cd concentrations of all samples were below detectable limit (0.01 mg/L) over the incubation process.

4. Discussion

4.1. Environment in incubation process

The simulated process in the MSWI bottom ash or mixtures with crushed incombustibles is illustrated in **Fig. 10**. The difference between the incubation process and the actual landfill process is that there is no leachate generated and no acid rain permeation in the former. So it is possible to evaluate ion and heavy metal solubility without thinking about the loss through leachate that usually happens at a landfill site and in a column test. CO_2 , O_2 and H_2O were supplied from the external environment, or were the products of the chemical and biological reactions. As shown in **Fig. 10**, the acidities are mainly from the dissolved CO_2 in water and the organic acids. In addition, sulfate and other sulphides such as H_2S were also conducive for the pH decrease. The pH of BA and MIX shifted from a high alkaline to a moderate level after a 1 year incubation process. With the pH change, the solubility of the heavy metals was also altered. In addition, some organic matter biodegradation was occurring. The soluble HSs were one of the main components of dissolved organic matter, and followed the same variation pattern as DOC. HSs have two different kinds of effects on the solubility of heavy metals. On the one hand, soluble HSs (mainly fulvic acid) promote the leachabilities of heavy metals binding with them. On the other hand, stable HSs (mainly humic acid) stabilize the heavy metals by complexing with them.

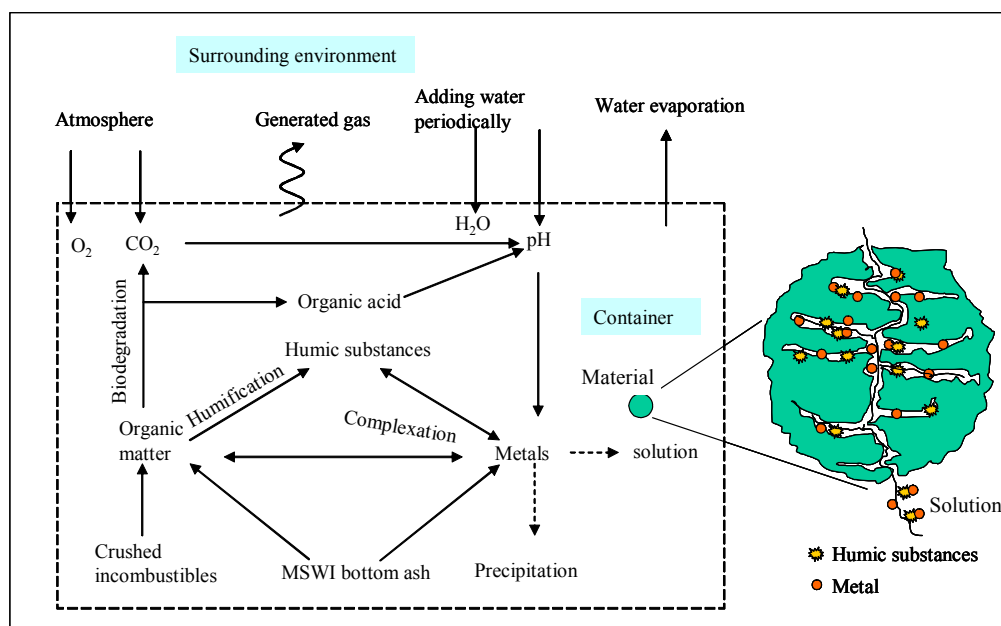


Fig. 10 Schematic illustration of incubation process.

4.2 Effect of pH on the leaching behavior of Cr, Zn, Pb and Cu

The solubility of heavy metals is often controlled by pH value. Carbonation process which can result in pH decrease has significant effect on the matrix dissolution or stabilization. The leaching concentration of Cr, Cu, Zn and Pb with pH is depicted in **Fig. 11**.

The leaching behavior of Cr has been well described⁷. Firstly, the release of chromium (VI) from the fresh MSWI bottom ash is controlled by chromate monophase ($\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{CaCrO}_4\cdot 15\text{H}_2\text{O}$) and chromate ettringite ($\text{Ca}_6\text{Al}_2(\text{CrO}_4)_3(\text{OH})_{12}\cdot 26\text{H}_2\text{O}$), and monophase could be replaced by ettringite when the pH decreased to around 11¹⁴⁻¹⁵. The stability of chromate ettringite is largely dependent on pH value and the concentrations of other competing oxyanions. At high pH (11.5-9.5), the increased sulfate and carbonate-ion compete with the chromate in precipitation or adsorption reactions, resulting in a higher Cr leaching concentration. In addition, ettringite began to dissolve when pH dropped to below 11, which lead to more Cr release¹⁵. With further decrease in pH, carbonate turned into bicarbonate, which does not form insoluble compounds. There was less competition for the chromate to precipitate or adsorption. So in neutral or acid conditions Cr was adsorbed to amorphous iron and aluminum oxides, and thereby its concentration decreased again.

The Cu concentration had an apparently strong correlation to pH value ($R^2 = 0.85-0.92$). Although Cu leachability seems to be controlled mainly by pH, the leachability might not depend entirely on pH. When large differences between pH values were noticed at 37 °C and 50 °C from the 24th to the 40th weeks, Cu concentrations showed no difference. And as fresh bottom ash has been previously studied using the pH depending test, Cu concentrations usually reach minimum value at pH 8 or 9, then increased with the pH decrease^{7, 16}. However, this phenomenon was not observed in the incubated bottom ash in this study. Precipitation of Cu with carbonate was one reason for the decrease of Cu concentration. Other factors must be considered to explain the Cu leaching behavior.

Pb leaching behavior during the incubation process is interesting. Leaching concentration was lower at pH 9-11 than that at other pH ranges. Pb is an amphoteric element, so it is soluble at high and low pH value. It was found that Pb leachability is controlled by the pH.

Leaching concentration of Zn was slightly increased at pH of 11-12. And it was lower than the detectable limit at other pH values.

The pH of CI leachate fluctuated in a narrow pH range during the incubation process. Hence the effect of pH on the leaching behavior of heavy metals from CI could not be observed.

4.3 Effect of DOC on the leaching behavior of Cr, Pb, Zn and Cu

The concentrations of VFA were relative stable at the early incubation process, and decreased dramatically when pH approached 9, and HSs had the same evolution pattern as DOC. We can assume that the effect of DOC on the leaching behavior of heavy metals will be the same as the effect of HSs. The relationship between the leaching concentrations of Cr, Pb, Zn and Cu and DOC from BA and MIX is shown in **Fig. 12**.

It seemed that the leaching concentrations of Cr and Cu had a positive correlation with the DOC, and this correlation became stronger with the increasing DOC concentration. On the other hand, DOC had less effect on their leachabilities of Zn and Pb. Although it has been postulated that Cu leachability is controlled by the complexation with organic matter^{17, 18}, strong relationship between DOC and Cu was not significant as had been expected at the start of this study. As shown in **Fig. 12**, the leaching concentration of Cu from MIX was higher under 37 °C with low DOC, while it was lower in the sample under 50 °C with higher DOC concentration.

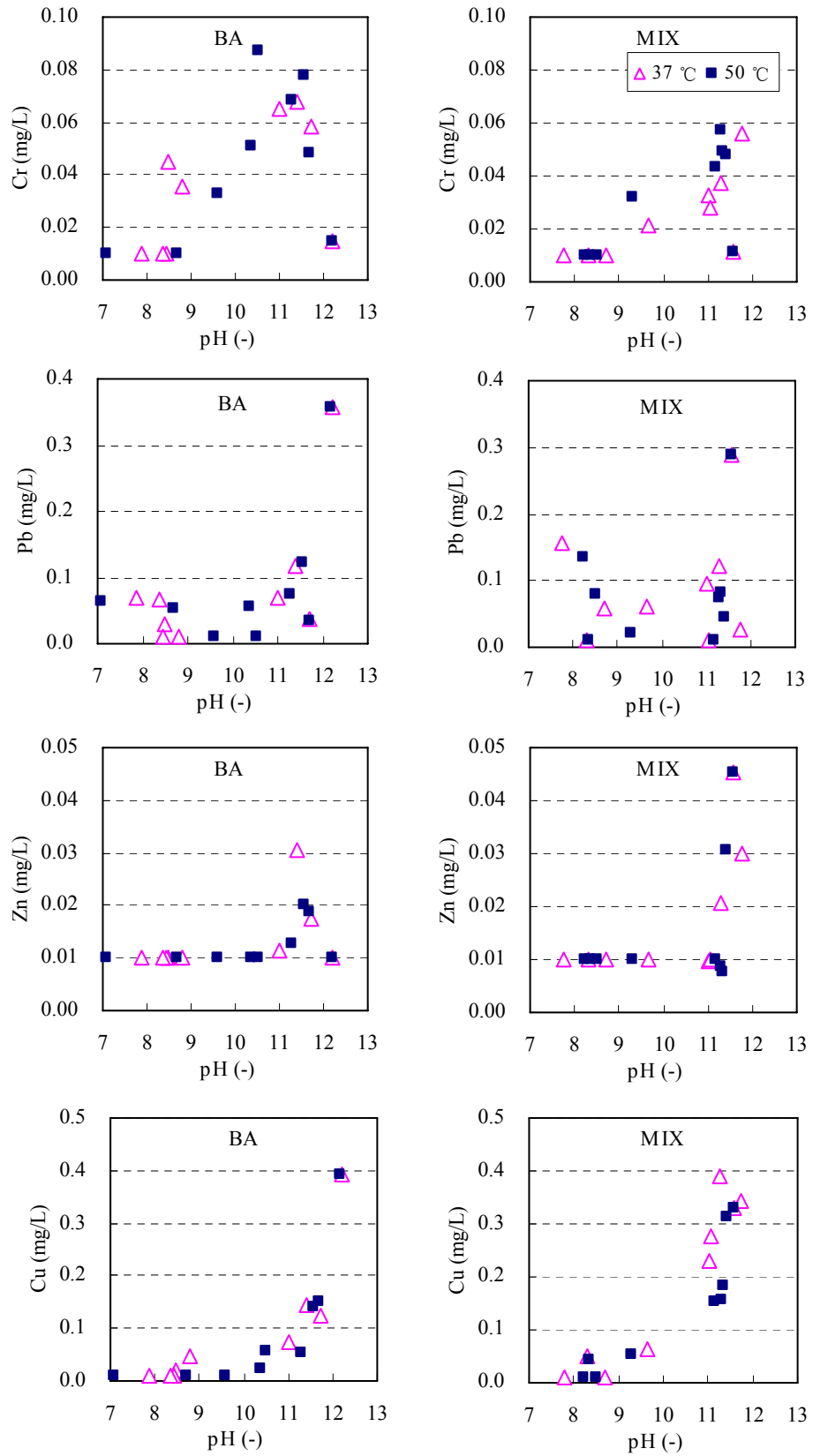


Fig. 11 Leaching concentrations of Cr, Zn, Pb and Cu as a function of pH.

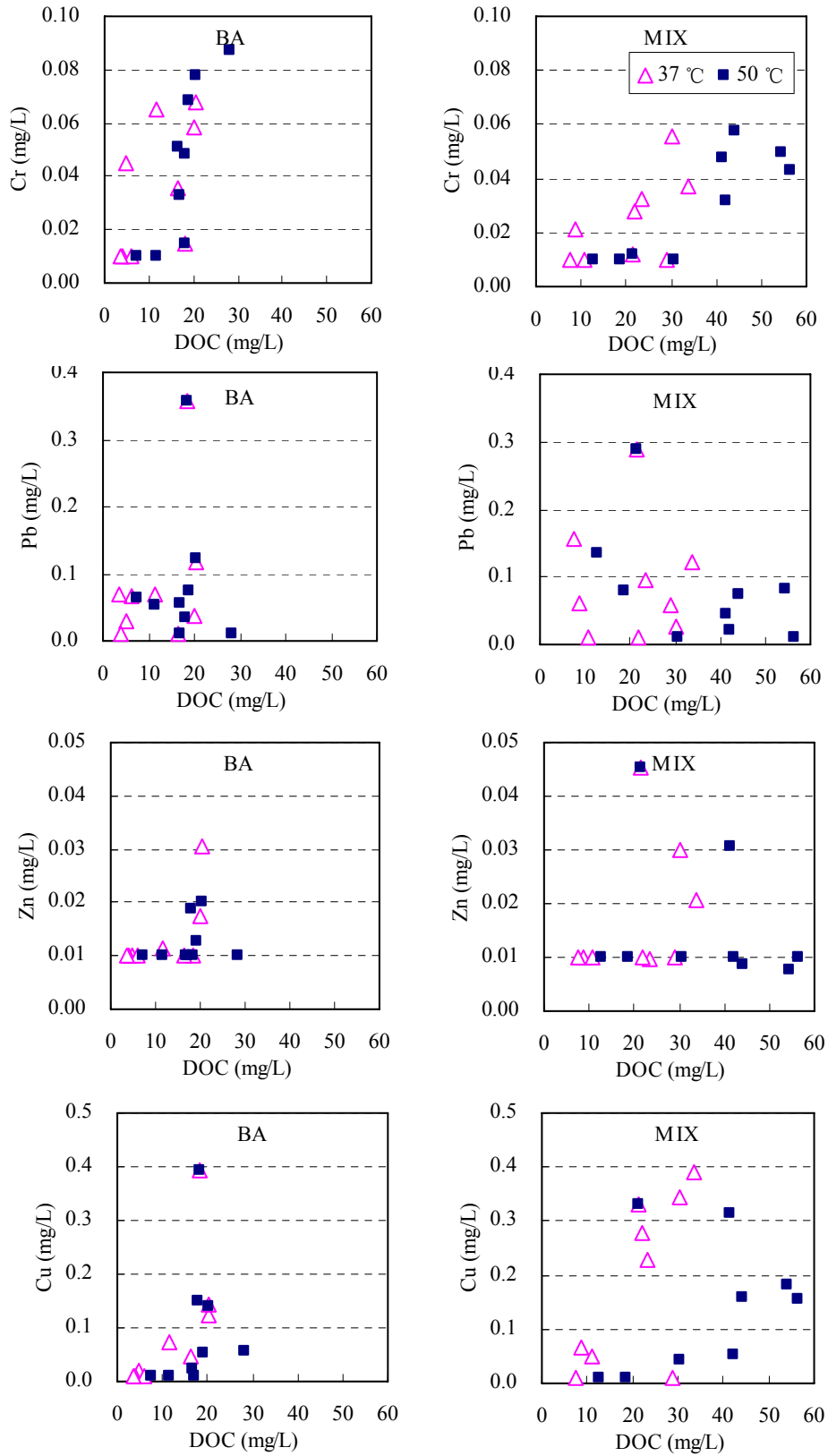


Fig. 12 Leaching concentrations of Cr, Pb, Zn and Cu as a function of DOC.

In the case of the CI incubated under 50 °C, higher DOC concentration corresponded to a higher leaching concentration of Pb, Zn and Cu (**Fig. 5** and **Fig. 9**). It was implied that dissolved organic matter could promote the leachabilities of these heavy metals from CI. MSWI bottom ash was a different matrix with CI or other material such as MSW and soil. Even MIX contained more dissolved organic matter than BA, which could not result in the more heavy metals leaching out. This indicated that bottom ash matrix had a stronger binding capacity to heavy metals, and other factor was important in controlling the leaching behaviors of heavy metals.

5. Conclusions

This study evaluated the soluble organic matter characteristics of bottom ash and the leaching behavior of heavy metals during an incubation process over a period of 1 year. DOC in the leachate from MSWI bottom ash was mainly composed of soluble HSs and VFA, and their concentrations fluctuated throughout the incubation period, indicating that biodegradation process had occurred during the incubation process. The pH of bottom ash decreased from high alkaline to moderate level and the release of Cr, Zn and Cu (exception of Pb) could satisfy the legal limit for soil pollution standard after 1 year incubation process. The ettringite formation and carbonation reaction were the main factors controlling the pH decrease, which further affects the leaching behavior of heavy metals. The correlation of dissolved organic matter with the leaching behavior of Cu was not significant as described in other studies. Although it can not disregard the fact that the CO₂ produced from bottom ash itself is enough to neutralize the bottom ash, i.e. that pH can decrease to neutral or acid condition even without atmospheric CO₂.

References

- 1) Annual Report on the Environment and the Sound Material-Cycle Society in Japan 2007, Source: Ministry of the Environment of Japan (2007)
- 2) S. Ferrari, H. Belevi and P. Baccini; Chemical speciation of carbon in municipal solid waste incinerator residues, *Waste Management*, Vol. 22, No.3, pp.303-314 (2002)
- 3) H. Zhang, T. Shimaoka and J.J. Lee; Variation of Organic Fractions in a Landfill mainly disposed with municipal solid waste incineration residues, *Proceedings of 4th Asian-Pacific Landfill Symposium*, Shanghai, China, pp.325-333 (2006)
- 4) F. H. Frimmel and L. Huber; Influence of humic substances on the aquatic adsorption of heavy metals on defined mineral phases, *Environment International*, Vol. 22, No. 5, pp. 507-517 (1996)
- 5) S. Bozkurt, L. Moreno and I. Neretnieks; Long-term processes in waste deposits, *The Science of the Total Environment*, Vol. 250, pp. 101-121 (2000)
- 6) J.M. Chimenos, A.I. Fernández, et al.; Short-term natural weathering of MSWI bottom ash as a function of particle size, *Waste Management*, Vol. 23, No. 10, pp. 887-895 (2003)
- 7) T. Van Gerven, E. Van Keer, et al.; Carbonation of MSWI-bottom ash to decrease heavy metal leaching, in view of recycling, *Waste Management*, Vol. 25 pp. 291-300 (2005)
- 8) J.J. Steketeer, R.F. Duzijn and J.G.P. Born; Quality improvement of MSWI bottom ash by enhanced aging, washing and combination processes, *Studies in Environmental Science*, Vol. 71, pp. 13-23, (1997)
- 9) C.A. Johnson and G. Furrer; Influence of biodegradation processes on the duration of CaCO₃ as a pH buffer in municipal solid waste incinerator bottom Ash, *Environment Science and Technology*, Vol. 36, No. 2, pp. 215-220 (2002)

- 10) H. Belevi, D.M. Stämpfli and P. Baccini; Chemical behavior of municipal solid waste incinerator bottom ash in monofills, *Waste Management & Research*, Vol. 10, No. 2, pp.153-167 (1992).
- 11) A.V. Zomeren and R.N.J. Comans; Speciation and characterization of organic carbon in Municipal Waste Incineration (MSWI) bottom ash, *WASCON 2003, Proceedings of the fifth international on the environmental and technical implications of construction with alternative materials*, pp.657-666 (2003)
- 12) J.A. Meima and R.N.J. Comans; Overview of geochemical processes controlling leaching characteristics of MSWI bottom ash, *Studies in Environmental Science*, Vol. 71, pp. 447-457 (1997)
- 13) J. Todorović and H. Ecke; Demobilisation of critical contaminants in four typical waste-to-energy ashes by carbonation, *Waste Management*, Vol. 26, No. 4, pp. 430-441 (2006)
- 14) C. Palmer; Precipitates in a Cr (VI)-contaminated concrete, *Environment Science and Technology*, Vol. 34, pp. 4185-4192 (2000)
- 15) M. Chrysochoou and D. Dermatas; Evaluation of ettringite and hydrocalumite formation for heavy metal immobilization: Literature review and experimental study, *Journal of Hazardous Materials*, Vol. 136, No. 1, pp. 20-33 (2006)
- 16) J.J. Dijkstra, H.A. Van der Sloot; The leaching of major and trace elements from MSWI bottom ash as a function of pH and time, *Applied Geochemistry*, Vol. 21, No. 2, pp. 335-351 (2006)
- 17) A.V. Zomeren and R.N.J. Comans; Contribution of natural organic matter to copper leaching from municipal solid waste incinerator bottom ash, *Environment Science and Technology*, Vol. 38, No. 14, pp. 3927 -3932 (2004)
- 18) S. Olsson, J.W.J. Van Schaik, et al.; Copper(II) binding to dissolved organic matter fractions in municipal solid waste incinerator bottom ash leachate, *Environment Science and Technology*, Vol. 41, No. 12, pp. 4286 – 4291 (2007)