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Solubility and adsorption characteristics of Pb in leachate from MSW incinerator bottom ash

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Abstract

The solubility and adsorption characteristics of Pb in leachate from municipal solid waste incineration (MSWI) bottom ash was studied by batch experiments. The solubility of Pb in leachate was 1–2 orders of magnitude lower than that of Pb in carbonate solutions. Adsorption experiments showed bottom ash have high adsorption capacity for lead. To evaluate a controlling component in bottom ash, quartz, Al and Fe powder as well as their ions were examined over wide ranges of pH at 25 °C. The results showed that only Fe powder had high adsorption capacities for Pb, indicating that iron oxides in bottom ash would be one of major adsorbent for Pb. Adsorption diagram, constructed by using the data of adsorption experiments, fitted the experimental data of Pb in leachate very well. It suggests that the decrease of Pb solubility in leachate is mainly due to adsorption reaction.

Keywords – lead; leachability; bottom ash; adsorption; iron oxides; solubility

1. Introduction

Municipal solid waste incineration (MSWI) plays an important role in Japan where landfill sites are difficult to be obtained. Approximately 74% of total MSW is combusted in about 1854 incineration plants (Sambongi et al. 1996). In Japan, as in other countries, fly ash for landfill disposal should undergo an intermediate treatment for toxic matter control such as dioxins and environmentally relevant metals, while bottom ash comes to landfill disposal directly.

However, bottom ash contains considerable concentrations of lead, which is amphoteric and strongly pH-dependent (Sawell et al. 1988; Van der Sloot et al. 1989; Stegemann et al. 1995). In this regard, potential hazards still exist in landfill where the chemical conditions (i.e., pH, Total Inorganic Carbon (C_T), Redox etc.) are apt to change significantly with time. Kersten et al. (1997) reported that the lead concentrations in leachate from a MSWI bottom ash monofill were a few orders of magnitude lower than solubilities of the Pb oxide or carbonated phases. Similar trends were observed in a batch experiment (Johnson et al. 1996) and column experiments (Seino et al. 1999; Sekito et al. 2000). However, little literature is available on the controlling mechanisms or factors effecting on the solubility of lead in leachate from bottom ash.

The objectives of this study were (1) to examine the solubility of Pb in leachate of MSWI bottom ash by batch tests over a wide range of pH, (2) to determine the controlling mechanisms or factors effecting on the solubility of Pb in leachate from bottom ash

2. Materials and methods

2.1 Sampling and sample preparation

Bottom ash used in batch tests was obtained from S-K facility near Sapporo city, Japan. This

mass burn system (stoker type) incinerates approximately 120 ton/day of municipal solid waste, under an incineration temperature of approximately 850 °C. Prior to experiments, coarse particles, larger than about 50mm diameter, were separated from bottom ash manually. The sample sieved through a 5.6 mm sieve was dried on aluminum plates at 90 °C–110 °C for 24 h. This dried sample was ground for 12 h in a ball mill to obtain representative samples (95% of mass below 125 µm) for batch tests. Samples were stored at 20 °C–25 °C. The composition of the sample is summarized in Table 1.

2.2 Methods

2.2.1 Solubility of Pb carbonate and hydroxide phases (Test A)

Theoretical solubility model of Pb carbonate and hydroxide phases was developed through an extensive literature review and computations for equilibrium constants of interest. Table 2 is a list of the pertinent equilibrium reactions for Pb used in this study. Equilibrium constants are also listed with reference sources. Although the data is not shown, hydrocerussite ($\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2(\text{s})$) species was identified in the ranges of pH 7.8–11 by X-ray diffraction analyses, as has been found in other studies (Marani et al. 1995, Patterson 1982). The solubility product of $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2(\text{s})$ was computed using the relationships $\Delta G^\circ = -RT \ln K$. Table 2 presents standard state free energy of formation (ΔG°) data of $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2(\text{s})$ equilibrium reaction. Log equilibrium constant of $\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2(\text{s})$ computed above was used in this work instead of Patterson's value (Table 2).

In order to ascertain the theoretical values in equilibrium with lead carbonates and hydroxide, a series of batch solubility experiments were performed at 25 ± 0.3 °C. 0.2 g PbCl_2 reagent was added into a glass flask containing 200 mL of distilled water. pH adjustments were made with NaOH or HNO_3 to achieve desired values. For Total Carbonate Concentration (C_T) adjustment, weighted amounts of sodium carbonate (Na_2CO_3) were added and set at about C_T 10 mg/L. pH was readjusted and controlled for the desired value for 30 minutes. The samples were filtered through 0.45 or 0.1 µm membrane filters

2.2.2 pH-Dependent leaching (Test B)

A series of batch leaching experiments were carried out to obtain information on the solubility of Pb in leachate over a wide pH range with various reaction times. The procedure of batch experiments is as follows: 2 g (dry weight) of pulverized bottom ash were mixed with 200 mL of distilled water in a glass flask at 25 ± 0.3 °C. pH adjustments were made with HNO_3 or NaOH to achieve desired values, and then kept stirring slowly for 30 min, 2 h, 4 h and 24 h, respectively.

2.2.3 Adsorption studies (Test C)

Adsorption isotherm experiments of bottom ash (Test C1) were conducted as follows: 0.1 g (dry weight) of pulverized bottom ash was added in 100 mL of distilled water. After pH adjustment was made to 6, 8 or 10, varying amounts of lead nitrate were added and stirred for 30 min. A repeat experiment without bottom ash was also conducted to obtain the solubility of Pb in leachate at pH 6, 8 and 10 for a reference.

Also, in order to examine adsorption of Pb to major constituents (Si, Al, and Fe) in bottom ash, the following experiments (Test C2) were conducted: The lead initial concentration was set at 1 mg/L. It is called 'control solution'. Then, weighted amount of SiO₂ powder, Al powder or Fe powder which are major components in bottom ash (Table 1) or some Na₂SiO₃, Al(NO₃)₃, or Fe(NO₃)₃ solution was added to the control solution and stirred slowly for 30 min.

Furthermore, adsorption isotherm experiments of Fe powder (Test C3) were conducted to examine its adsorption characteristics of Pb: Ash extraction solution was previously arranged by stirring 1g-ash/L-distilled water at three different pH values (6, 8 and 10) for 30 min. Then, various amounts of lead nitrate were added in 100mL of each pH extraction solution. pH of the solution was adjusted to preset value (pH 6, 8 or 10) just prior to adding 0.01 g Fe powder. HNO₃ or NaOH was used for pH adjustment and room temperature (25 ± 0.3 °C) controlled throughout the experimental period.

2.2.4 Analytical

All samples were filtered through a 0.45 μm (when needed, 0.1 μm) membrane filter and measured pH and total inorganic carbon (C_T). pH was measured by a pH meter (HORIBA B212). pH values were reported to the nearest 0.01 pH unit, and total inorganic carbon (C_T) concentrations were determined by TOC analyzer (SHIMADZU TOC-5000). At the same time, some portion of the filtered sample was acidified with HNO₃ (pH < 2) for analysis of heavy metals. They were determined by a graphite furnace atomic absorption spectrophotometer (HITACHI Z-8200).

3. Results and discussion

3.1 Comparison of the theoretical & experimental solubility of Pb (Test A)

The results for lead solubility experiments at about C_T 10 mg/L are plotted in Fig. 1. The theoretical solubility curves with Pb₃(CO₃)₂(OH)₂(s) species present & absent at C_T 10 mg/L are also presented for a reference. Experimental data (Test A) were in good agreement with the theoretical solubility curve with Pb₃(CO₃)₂(OH)₂(s) present. Thus, the solubility curve of Pb with Pb₃(CO₃)₂(OH)₂(s) species present was used to compare with experimental data obtained in this study for a reference. Deviations from the curve were occurred at the ranges of pH 9 to

10 in the case of 0.45 μ m membrane filtration, as was observed in Patterson's work (1982). The results of 0.1 μ m membrane filtration suggest that the deviation should be attributed to precipitates size on solubility.

3.2 Comparison of column and batch leaching experiments (Test B)

By column experiments conducted until 3 liquid to solid (L/S) ratios, Seino et al. (1999) showed that Pb concentrations in leachate are mainly controlled by leachate pH values which resulted from different CO₂ gas contact times with bottom ash (Fig. 2a). Fig. 2b (Test B) shows the relations between pH and lead concentrations by batch leaching test (100 L/S ratios). As shown in Fig. 2b, 30 min of reaction time seems to be sufficient to reach equilibrium in this experiment.

From the results of the column and the batch leaching experiments, leachate pH had a notable impact on the solubility of lead, but L/S ratios and contact times between the leachant and the particles appeared to be little influence on the solubility of lead.

Interestingly, the leaching trend of Pb in the batch tests was similar to the column test results and lead concentrations from column and batch leaching experiments were all about 1–2 orders of magnitude lower than the theoretical solubility curve over a wide range of pH. It suggests that inherent physico-chemical property of bottom ash should act on the leaching behaviour.

3.3 Adsorption isotherms between bottom ash vs. lead (Test C1)

The results of adsorption isotherm experiments for Pb at fixed pH are plotted in Fig. 3. The adsorption data were fitted to Henry's isotherm very well. Table 4 presents aqueous equilibrium concentrations of lead with/without bottom ash adsorbent at pH 6, 8 and 10. The amount of Pb adsorbed on bottom ash, or Q_e was obtained by the following relations;

$$Q_e(\text{mg} / \text{g} - \text{ash}) = ([Pb]_i + [Pb]_l - [Pb]_s) / S$$

where $[Pb]_i$ = amount of Pb added initially (mg); $[Pb]_l$ = amount of Pb leached from bottom ash at a specific pH (mg); $[Pb]_s$ = amount of soluble Pb in solution (mg); S = weight of bottom ash. $[Pb]_l$ was obtained in batch leaching tests (Test B).

As shown in Fig. 3, aqueous equilibrium concentrations of Pb are initially in proportion to the amount of Pb adsorbed on bottom ash; after adsorption capacity is saturated, the equilibrium is determined by the solubility of Pb hydroxide or carbonated phases. From this result, adsorption reaction is considered to be the predominant mechanism controlling the solubility of Pb in leachate because leachable concentrations from bottom ash is much less than the adsorption capacity of bottom ash (about 10–20 mg-Pb/g-bottom ash). Then, what is a controlling component (adsorbent) in bottom ash? Bottom ash contains abundant silicon, aluminum and

iron as presented in Table 1. According to other studies, silicon, aluminum, and iron do not volatilize under the prevailing oxidizing conditions. So most of them are transferred to bottom ash (Belevi et al. 2000a; Belevi et al. 2000b; Verhulst et al. 1996). For these reasons, the amount of Si, Fe and Al corresponding to that of bottom ash was applied to the next experiment.

3.4 Effect of Si, Al, and Fe components on Pb solubility (Test C2)

Fig. 4 shows the trend of aqueous concentrations of Pb at various pHs, when added with various concentrations of Na_2SiO_3 or SiO_2 powder. In addition, control solution contained the initial lead concentration (1 mg-Pb/L) is also plotted as a reference. As shown in Fig. 4, aqueous concentrations of lead didn't decrease regardless of the amounts of SiO_3^{2-} added, compared to the control solution. Although several other silicon-containing minerals exist in bottom ash, Quartz (SiO_2 powder), rich in bottom ash, also didn't appear to interact with soluble Pb in water.

Likewise, aqueous concentrations of lead contained various $\text{Al}(\text{NO}_3)_3$ or Al powder are plotted in Fig. 5. It is similar to the results of Si experiments except for 5 mg- Al^{3+} /L at pH 8, but this high ratios of Al to Pb concentration is not likely to exist in leachate from bottom ash. Therefore, Al component in bottom ash doesn't seem to strongly effect on the solubility of Pb in leachate from bottom ash. These observations are in support of earlier studies which also found that Pb adsorption by Al oxides was negligible (Deming Dong et al. 2000; Jennifer et al. 1995; Nadya et al. 1999).

The experimental data of Fe powder and $\text{Fe}(\text{NO}_3)_3$ are plotted with those of control solution in Fig. 6. It appears that Fe powder is an effective component for decreasing the solubility of Pb at pH 6, 8 and 10.

3.5 Adsorption isotherms between Fe powder vs. lead (Test C3)

Adsorption isotherms for Pb by Fe powder at pH 6, 8 and 10 are presented in Fig. 7. The experimental data were in good agreement with Henry's isotherm until maximum adsorption capacity of Fe powder reached. After that, it was controlled the solubility of Pb hydroxide or carbonated phases, as found the same way in the adsorption isotherm experiments with bottom ash. $\text{Fe}(\text{OH})_3(\text{s})$ precipitates had the maximum adsorption capacity of 1–2 mg-Pb/g- $\text{Fe}(\text{OH})_3$ in this work (data not shown). Thus, the contribution of Pb adsorption on $\text{Fe}(\text{OH})_3(\text{s})$ precipitates is relatively small, compared to Fe powder (50–300 mg-Pb/g-Fe powder). Fig. 7 showed that Fe powder had high adsorption capacity for Pb at pH 6, 8, 10.

During the combustion process, Fe oxides, very stable at high temperature, become concentrated on the bottom ash forming a surface coating. So, the surface coating of amorphous iron oxide is considered to exert the primary controls on lead in water. Indeed, a number of researchers have reported that Fe oxides have a high adsorption capacity for lead in aquatic

environmental systems (Deming Dong et al. 2000; Edwards et al. 1989; Jennifer et al. 1995; Nadya et al. 1999; Smith, E.H. 1996; Swallow et al. 1980; Theis et al. 1992). Therefore, considering the combustion process of MSW and the characteristics of Fe oxide, it suggests that iron-bearing bottom ash should be one of the major controlling components with regard to lead in water.

However, considering Fe fraction (4–5%) of bottom ash (Table 1) and its maximum adsorption capacity (Fig. 7), a single component of iron fraction in bottom ash can't explain completely the adsorption capacity of bottom ash obtained in this work (Fig. 3). This disparity may arise from differences in particles size and a surface area between Fe powder and the metal iron fraction in bottom ash used in this study. Otherwise, other components in bottom ash, such as manganese oxides (Fan et al. 1995) or soluble fulvic acid (Saar et al. 1980), may partly contribute to the adsorption of Pb.

3.6 Pb solubility diagram based on adsorption reaction (Adsorption diagram)

Using adsorption data between bottom ash and Pb in Fig. 3, adsorption diagrams in Fig. 8 are constructed by the following equations.

Mass balance relationships for lead in solution:

$$S \times Pb(a) + W \times Pb(aq) = Pb(i) \times S \quad (1)$$

where S = weight of bottom ash (g)

W = amount of distilled water (L)

$Pb(a)$ = the amount of lead adsorbed per g of bottom ash (mg/g)

$Pb(aq)$ = equilibrium concentrations of lead in solution (mg/L)

$Pb(i)$ = the amount of lead leached per g of bottom ash at a specific pH value (mg-Pb/g-bottom ash)

The form of Henry's isotherm equation is:

$$Pb(a) = k \times Pb(aq) \quad (2)$$

where k (L/g) is distribution coefficients as a function of pH.

distribution coefficients, k can be determined from three slopes of adsorption lines at pH 6, 8 and 10 in Fig. 3.

Using three different distribution coefficients at pH 6, 8, and 10, respectively, correlation between k and pH is governed by the following equation:

$$\log k = -0.1728(pH)^2 + 3.0014(pH) - 11.093 \quad (3)$$

where the slope at pH 6, 8 and 10 was 5.0, 73, and 44, respectively.

By substituting $k \times Pb(aq)$ for $Pb(a)$ in equation (1), the following equation can be obtained:

$$Pb(aq) = \frac{Pb(i)}{k + W/S}$$

or, $Pb(i) = (k + W/S)Pb(aq)$ (4)

On the basis of the above equations, adsorption diagrams can be constructed as shown in Fig. 8. However, it is too difficult to obtain the amount of lead leached from bottom ash ($Pb(i)$) at a specific pH, since adsorption reaction occurs immediately after lead is leached from bottom ash. For that, adsorption diagrams were plotted with assumed $Pb(i)$ values; 0.1 mg/g, 1 mg/g, and theoretical solubility values of Pb carbonate and hydroxide phases at C_T 5mg/L. Fig. 8 showed adsorption curves fit the experimental data very well. This suggests that as a result of adsorption reaction, aqueous equilibrium concentrations of lead in leachate from bottom ash are 1–2 orders of magnitude lower than the theoretical solubility of Pb carbonate and hydroxide phases. In addition, it can be referred that the amount of lead leached from bottom ash, $Pb(i)$ in this experiment will be about 0.1 mg/g-ash in the range of pH 6–12, but increased with decreasing pH.

4. Conclusions

The following major results were obtained:

- (1) The results of batch leaching tests of bottom ash showed that equilibrium concentrations of Pb in leachate are 1–2 orders of magnitude below the theoretical solubility of Pb carbonate and hydroxide phases over a wide range of pH.
- (2) Bottom ash has a considerable adsorption capacity (10 to 20 mg-Pb/g-bottom ash) at pH 6, 8 and 10. Additionally, it was observed that equilibrium concentration of lead in leachate depends initially on the adsorption capacity of bottom ash; when the adsorption capacity reaches to saturation, the equilibrium depended only on the solubility of Pb carbonate and hydroxide phases. These trends were also observed in adsorption experiments with Fe powder.
- (3) Considering that the amount of leachable concentrations from bottom ash is very small, the solubility of lead in leachate would be controlled by adsorption reaction rather than by the solubility of Pb carbonate and hydroxide phases.

- (4) Fe powder showed high adsorption capacity for Pb at pH 6, 8, 10 unlike Al and quartz. Thus, iron-bearing, especially forming oxides, bottom ash is considered to be one of major adsorbent for lead in leachate.
- (5) Thus, the deviations from the lead solubility curve are considered to be due to adsorption reactions between iron oxides in bottom ash and lead in water.

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Table 1. Composition of MSWI bottom ash

Element	Unit (by weight)	Bottom ash ^a
Ca	%	15.1
Si	%	12.5
Al	%	6.31
Fe	%	4.02
Na	%	1.76
Mg	%	1.74
K	%	1.44
Cu	ppm	6740
Zn	ppm	4210
Pb	ppm	2650
Mn	ppm	895
Cr	ppm	158
Cd	ppm	1.8
IL ^b	%	7.5

^a HNO₃/HCl total digestion/AAS^b Ignition Loss at 600 °C for 3 hrTable 2. Equilibrium reactions for lead in the system Pb²⁺ - CO₂ - H₂O

Equilibrium Reactions	Log Equilibrium Constants at 25	Reference
1. Equilibrium among solutes and CO ₂ (g)		
H ₂ O = H ⁺ + OH	- 14	Stumm et al. (1996)
H ₂ CO _{3(aq)} = H ⁺ _(aq) + HCO _{3⁻(aq)}	- 6.3	Stumm et al. (1996)
HCO _{3⁻(aq)} = H ⁺ _(aq) + CO _{3²⁻(aq)}	- 10.3	Stumm et al. (1996)
2. Solid-solutions Equilibrium		
Pb(OH) ₂ (s) = Pb ²⁺ + 2OH	- 14.9	Patterson (1982)
Pb ²⁺ + OH = PbOH ⁺	6.3	Smith et al (1976)
Pb ²⁺ + 2OH = Pb(OH) ₂ ⁰	10.9	Smith et al (1976)
Pb ²⁺ + 3OH = Pb(OH) ₃	13.9	Smith et al (1976)
Pb(CO ₃) ₃ (s) = Pb ²⁺ + CO _{3²⁻}	- 12.8	Patterson (1982)
Pb ²⁺ + CO _{3²⁻} = PbCO ₃ ⁰	6.2	Stumm et al. (1996)
Pb ²⁺ + 2CO _{3²⁻} = Pb(CO ₃) ₂ ²⁻	9.4	Stumm et al. (1996)
Pb(CO ₃) ₂ (OH) ₂ (s) + 2H ⁺ = 3Pb ²⁺ + 2CO _{3²⁻} + 2H ₂ O	- 18.8	Patterson (1982)
Pb(CO ₃) ₂ (OH) ₂ (s) + 2H ⁺ = 3Pb ²⁺ + 2CO _{3²⁻} + 2H ₂ O	- 17.7	In this work

Table 3. ΔG° values for each chemical species

Species	ΔG° (kcal/mol)	Reference
Pb(CO ₃) ₂ (OH) ₂ (s)	407.3	Taylor et al. (1984)
H ⁺ _(aq)	0	Benfield et al. (1982)
Pb ²⁺ _(aq)	- 5.81	Benfield et al. (1982)
CO _{3²⁻} _(aq)	- 126.17	Benfield et al. (1982)
H ₂ O(l)	- 56.68	Benfield et al. (1982)

Table 4. Equilibrium soluble concentrations with/without bottom ash adsorbent

PH	Bottom ash			Without bottom ash (in extraction solution ^a)
	Ci (mg/L)	Ce (mg/L)	Qe (mg/g)	Cs (mg/L)
6	0.1	0.002	0.099	
	0.5	0.009	0.492	
	1	0.019	0.983	
	2.5	0.111	2.391	
	5	0.326	4.676	
	10	1.307	8.695	8.608
	25	4.078	20.922	
	30	8.486	21.515	28.58
	35	11.267	23.733	
	40	17.723	22.277	
8	0.1	0	0.1	
	0.5	0.0006	0.499	
	1	0.0013	0.999	
	2.5	0.0069	2.493	
	5	0.0277	4.972	
	8	0.0851	7.915	
	10	0.1114	9.889	0.1966
	13	0.1623	12.838	
	14	0.1858	13.814	
	16	0.1853	15.815	
10	20	0.1866	19.813	
	30	0.2082	29.792	0.1797
	0.1	6.59E-05	0.101	
	0.5	0.0037	0.497	
	1	0.0077	0.993	
	2.5	0.0233	2.478	
	5	0.0497	4.950	
	6	0.0454	5.955	
	7	0.1119	6.888	
	8	0.1066	7.893	
10	9	0.1185	8.882	
	10	0.1797	9.821	0.4382
	12	0.2224	11.777	
	14	0.32	13.68	
	16	0.3148	15.685	
	20	0.3394	19.661	
	30	0.2702	29.731	0.3387

^a denotes solution obtained with 0.45 μ m membrane filter after stirred 1g-ash/L-distilled water for 30min, Ci = initial lead concentration added, Ce = total lead concentration at equilibrium, Qe = amount of lead adsorbed on bottom ash, and Cs = experimental solubility values of Pb in extraction solution.

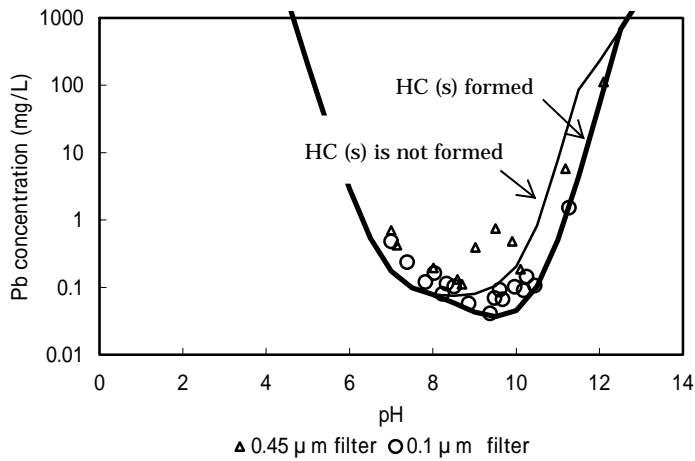


Fig. 1. Comparison of the theoretical and experimental solubility of Pb at C_T 10 mg/L. The bold line represents the theoretical solubility of Pb with $Pb(CO_3)_2(OH)_2$ species present at C_T 10 mg/L, HC stands for Hydrocerrucite ($Pb(CO_3)_2(OH)_2$).

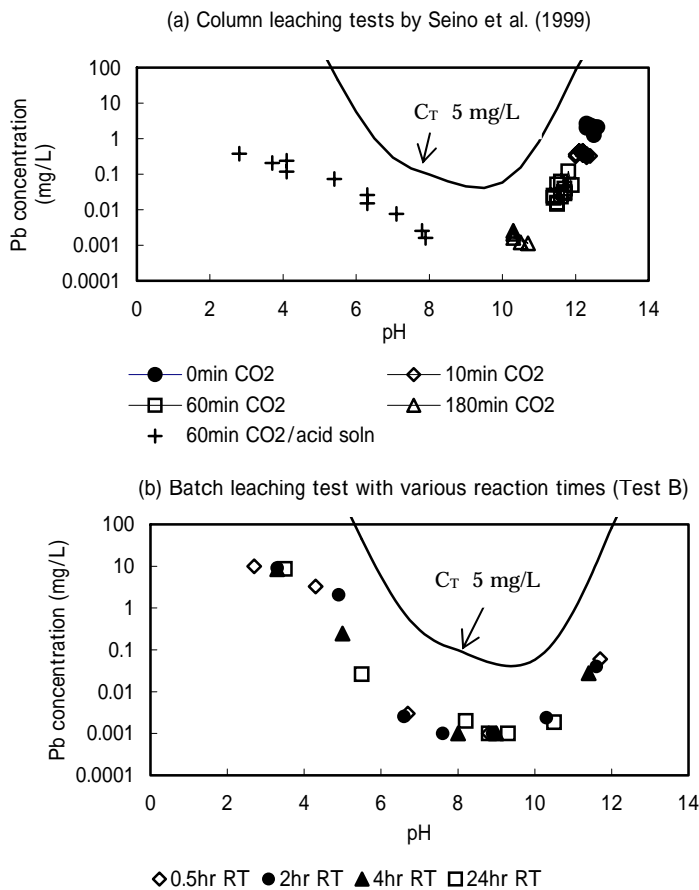


Fig. 2. Comparison of Pb concentrations in leachate from column and batch tests as a function of pH. The line represents the theoretical solubility of Pb at C_T 5 mg/L. '0 min CO₂' means bottom ash contacted with CO₂ gas for 0 minute. 'acid soln' is pH 2 solution and 'RT' reaction time.

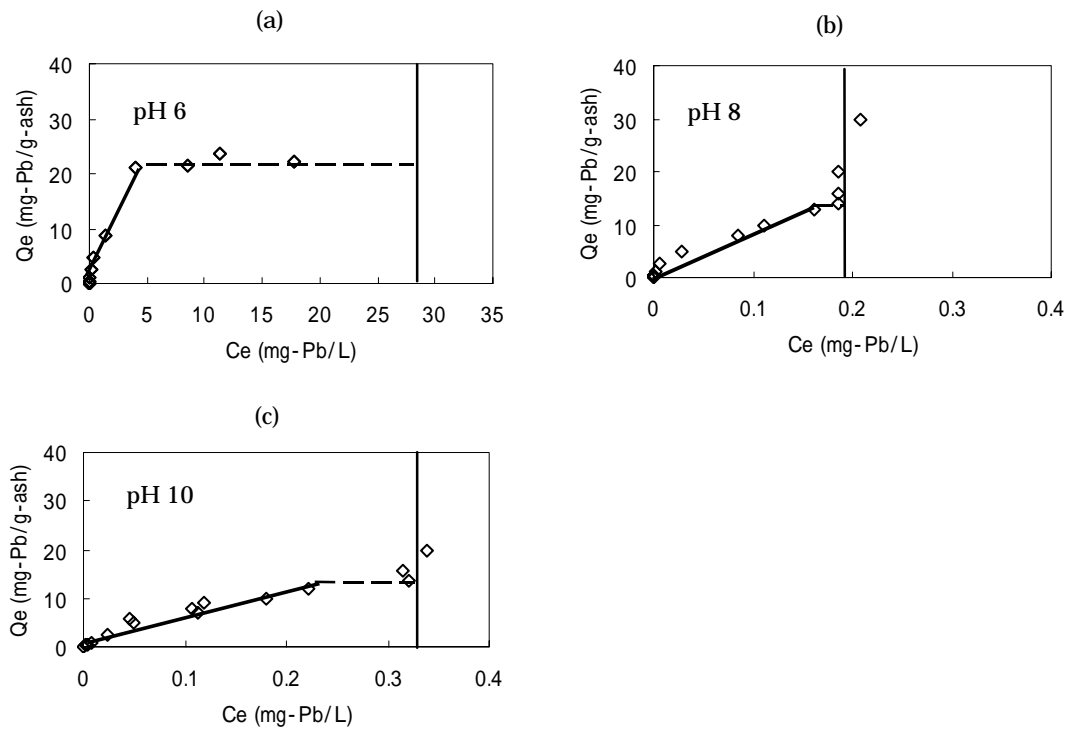


Fig. 3. Adsorption isotherms of Pb on bottom ash at pH6, 8 and 10. Pb^{2+} dosage ranges from 0.1 to 40 mg/L, — is adsorption line, - - - saturation line, and — experimental solubility line of lead (Test C1).

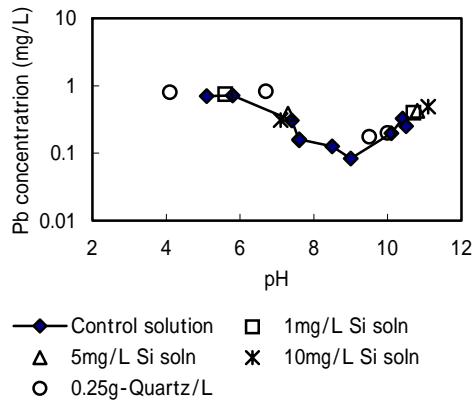


Fig. 4. Aqueous concentrations of Pb in the presence of various concentrations of Si or Quartz powder (Test C2).

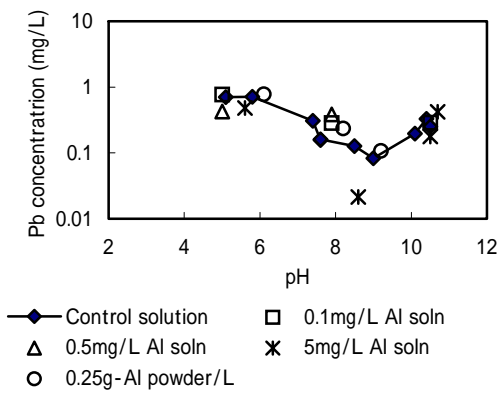


Fig. 5. Aqueous concentrations of Pb in the presence of various concentrations of Al or Al powder (Test C2).

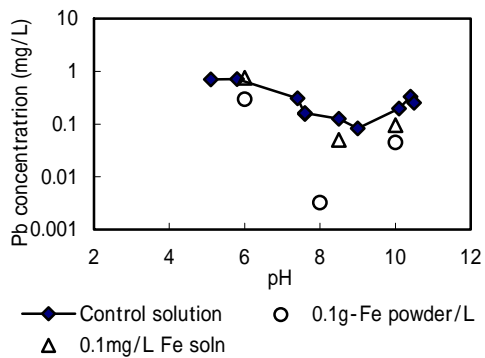


Fig. 6. Aqueous concentrations of Pb in the presence of Fe solution, or Fe powder (Test C2).

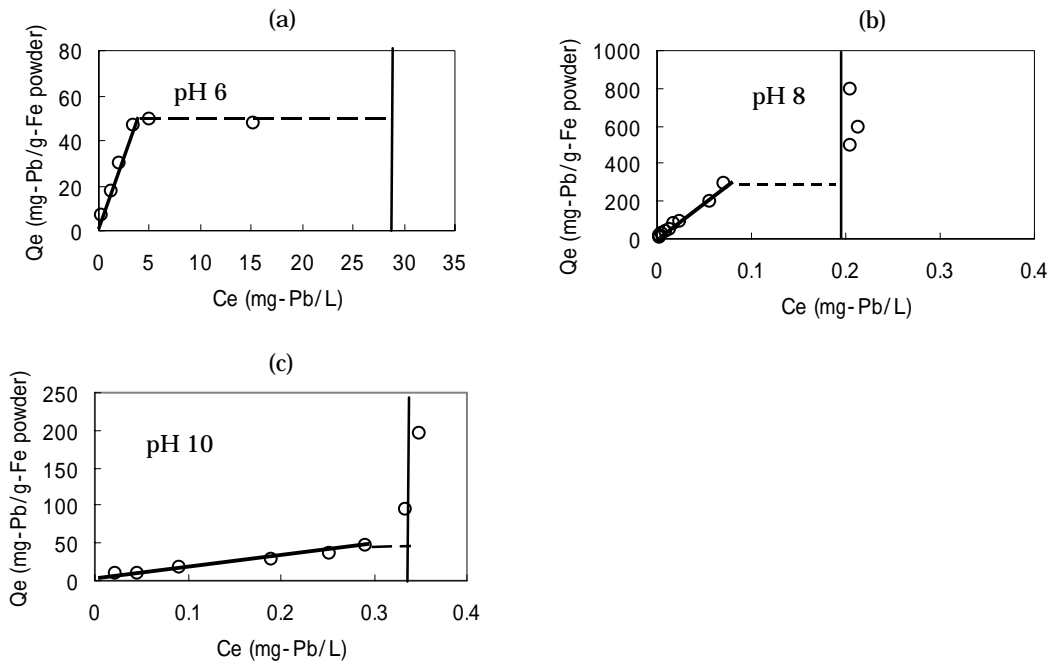


Fig. 7. Adsorption isotherms of Pb on Fe powder at pH6, 8 and 10. Pb^{2+} dosage ranges from 0.1 to 80 mg/L, — is adsorption line, - - - saturation line, and — experimental solubility line of lead.

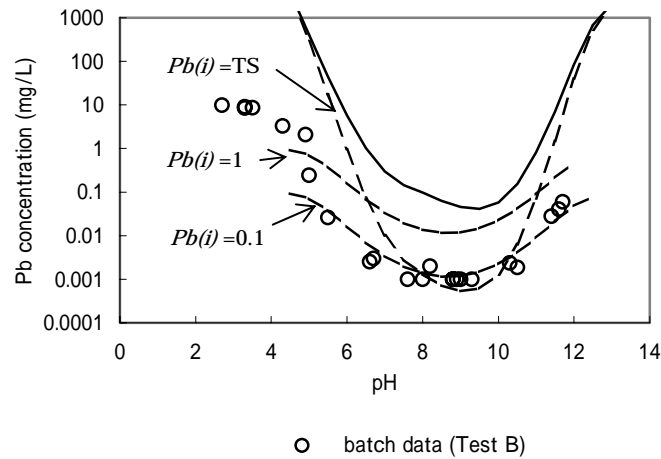


Fig. 8. Predicted adsorption diagrams at 0.1 mg-Pb/g-ash, 1 mg-Pb/g-ash, and the theoretical solubility values of Pb at C_T 5 mg/L as $Pb(i)$. — the theoretical solubility curve of Pb at C_T 5 mg/L, TS stands for theoretical solubility values.