Behavior of Heavy Metals in Landfilled Fly Ashes for 27 years

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1. INTRODUCTION

Incineration has played an important role in solving the scarcity of available land to accommodate the accumulated municipal solid waste (MSW). In 2020, 41.67M tons of MSW were generated, 79.1% of which were incinerated and converted to 3.64M tons of landfill waste (Ministry of the Environment, Government of Japan, 2022). MSWI produces a considerable amount of fly ash (FA) and bottom ash (BA), which contains harmful heavy metals such as Pb, Cd, etc. Generally, FA has a relatively higher heavy metal content than BA (Lu, et al., 2020). Therefore, MSWI FA needs to be pre-treated before landfill disposal according to the Waste Disposal and Public Cleansing Law in Japan (Sakai, 1996). There are far more landfill sites in mountainous areas than in other areas in Japan. The long-term stabilization of treated fly ash in mountainous landfills has not been clarified (Hirokawa & Fukuda, 2000).

The study investigates the behavior of fly ash from simulated mountainous landfill model columns for untreated, chelate-treated, and phosphate-treated fly ash, and slag. This study aims to determine whether landfilled FA has stabilized after 27 years. Column leaching experiment and Japan Leaching Test (JLT-13), were conducted on untreated, chelate-treated, phosphate-stabilized, and slag-treated FA.

2. MATERIALS AND METHODS

Ten landfill model columns were filled with different types of ash mix (Table 1). The top end of the column is exposed to natural rainfall, whereas the leachate is collected at the bottom. (Fig.1)

The BAFACG ash mix has a 60:20:15:5 ratio corresponding to BA: FA: Crushed Garbage: Compost. BAFA(U) and SMS have a 3:1 ratio for BA:FA whereas PMS has a 4:1 BA to FA ratio. The rest of the samples are pure FA and BA.

For JLT-13, 50g of ashes were collected from four collection points marked with 'x' in Fig. 1. First, 500mL of super pure water was added to achieve a liquid-to-solid (L/S) ratio of 10, then shaken for 6 h at 200 oscillations per minute. For the column experiment, 500 mL of leachate samples were collected. Next, the liquid samples of both tests were filtered through a glass-fiber filter, and 50mL was separated for analysis. Then, nitric acid was used to prepare the filtered samples for Inductively Coupled Plasma testing with 720 ICP-OES.

3. RESULTS AND DISCUSSION

3.1 Behavior of FA Leachate from Column Experiment

In a simulated environment where the ashes are exposed to natural rainfall, the amount of leached heavy metals for all samples, apart from slag-treated ash mix: SMS and PMS, in 1997 is significantly higher than that of 2002 as shown in Table 2(a). Slag-treated ash mix showed considerably low concentrations of heavy metals leached throughout the 27 years. By 2022, all five heavy metals meet the waste-water legal regulation in Japan of 0.1, 0.1, 2.0, 3.0, and 5.0 (mg/L) for Pb, Cd, Cr, Cu, and Zn, respectively (Yasuhara, 1995).

Table 1. Ash mix and treatment method

Column No.	Sample Name	Abbreviation
1	Untreated Bottom Ash, Fly Ash, Compost, Crushed Garbage	BAFACG(U)
2	Chelate treated BAFACG	BAFACG(C)
3	Phosphoric acid treated BAFACG	BAFACG(P)
4	Untreated Bottom Ash	BA(U)
5	Untreated Fly Ash	FA(U)
6	Untreated Bottom Ash + Fly Ash	BAFA(U)
7	Surface Melting Slag	SMS
8	Plasma Melting Slag	PMS
9	Chelate treated Fly Ash	FA(C)
10	Phosphoric acid treated Fly Ash	FA(P)

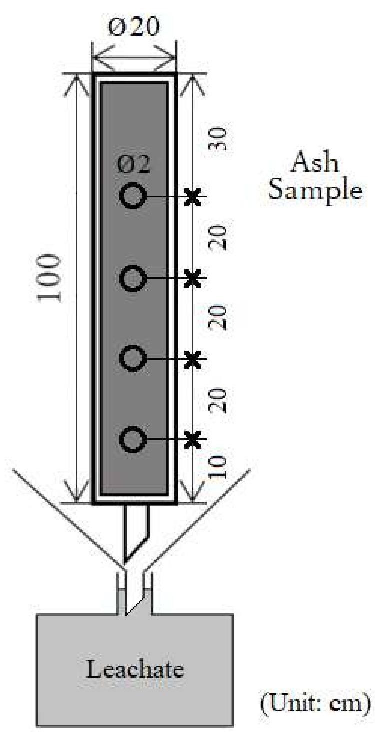

Fig. 1. Column Sketch

Table 2. Leached heavy metals concentration from (a) Column Leaching Experiment (b) Leaching Test (JLT-13)

(a)

#	Sample	Pb (mg/L)		Cd (mg/L)		Cr (mg/L)		Zn (mg/L)		Cu (mg/L)	
		1997	2022	1997	2022	1997	2022	1997	2022	1997	2022
1	BAFACG(U)	0.69	0.01	0.11	0.01	0.60	0.01	0.40	0.05	17.4	0.02
2	BAFACG(C)	0.45	0.01	0.23	0.01	0.40	0.01	0.90	0.02	5.00	0.01
3	BAFACG(P)	0.43	0.01	0.15	0.01	0.40	0.01	2.20	0.05	2.60	0.01
4	BA(U)	0.15	0.01	0.01	0.01	0.02	0.01	0.82	0.09	46.0	0.01
5	FA(U)	230	0.01	21.0	0.01	1.60	0.01	57.0	0.03	2.70	0.01
6	BAFA(U)	6.20	0.01	3.10	0.01	0.05	0.01	20.0	0.07	23.0	0.01
7	SMS	0.05	0.02	0.01	0.01	0.02	0.01	0.04	0.17	0.09	0.02
8	PMS	0.05	0.01	0.01	0.01	0.02	0.01	0.04	0.11	0.02	0.01
9	FA(C)	3.90	0.01	2.70	0.01	0.03	0.01	120	0.05	5.80	0.02
10	FA(P)	6600	0.01	75.0	0.01	0.02	0.01	3500	0.11	75.0	0.01

(b)

#	Sample	Pb (mg/L)		Cd (mg/L)	
		2000	2022	2000	2022
1	BAFACG(U)	0.07	0.04	0.01	>0.01
2	BAFACG(C)	0.49	0.04	>0.01	>0.01
3	BAFACG(P)	0.37	0.02	0.02	>0.01
4	BA(U)	0.05	>0.01	>0.01	>0.01
5	FA(U)	23.5	0.02	>0.01	>0.01
6	BAFA(U)	0.05	0.01	>0.01	>0.01
7	SMS	>0.05	0.02	>0.01	>0.01
8	PMS	>0.05	0.01	>0.01	>0.01
9	FA(C)	0.16	0.01	>0.01	>0.01
10	FA(P)	>0.05	0.01	>0.01	>0.01

Fig 2 shows a simulation of the relationship between Zn and pH where the Zn concentration changes is most drastic between pH value of 8.0 and 9.0. The Zn in all samples except FA(C) and a few outliers had a relatively low concentration of less than 0.08mg/L for 9 years up until 2011 (Fig 3(a)), before increasing in a random pattern for the next 10 years. Despite that, the Zn value remained below Japan's regulatory criteria for waste-water leachate.

The experimental data collected from 2003 to 2011 (Fig 3(a)) and 2012 to 2022 (Fig 3(b)) shows a similar trend when compared with the simulation graph in Fig 2, with a few outliers which may have been caused by experimental errors. The peak pH value of the leachate decreased from 11.2 (Fig 3(a)) to 9.7 (Fig 3(b)).

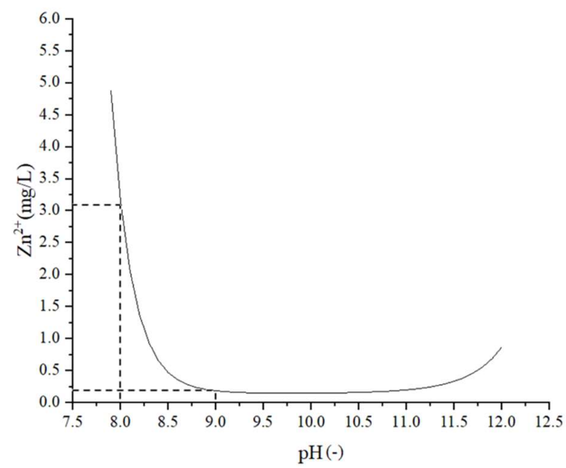

Fig 2. Simulation of Zn vs pH graph by Visual Minteq

3.2 Leaching Behavior of FA (JLT-13)

In 2000 (Table 2(b)), the leached Pb was relatively higher for samples containing a high FA ratio, while others had values below the machine detection limit. As for leached Cd in the same year, almost all samples are below the machine detection limit of 0.01 mg/L. In 2022, all the samples had leached Pb value of less than 0.04 mg/L and leached Cd value below the detection limit of 0.01 mg/L. Both heavy metals Pb and Cd satisfy the Japanese regulatory criteria for landfilling at 0.3mg/L (Kim, et al., 2003).

4. CONCLUSIONS

In conclusion, all heavy metals in the column experiment meet Japan's waste-water legal regulation of 0.1 mg/L for Pb and Cd, 2 mg/L, 3 mg/L and 5 mg/L for Cr, Cu, and Zn, respectively. The value of leached Zn is heavily dependent on the pH value of the leachate when the pH value falls between 8.0 and 9.0. Even though the heavy metals in all samples have very low leached values, further tests, such as a pH-dependent test are required to determine whether these heavy metals leach the same amount at different pH values to conclude that it has stabilized confidently. The leached Pb and Cd from the leaching test (JLT-13) fell way below Japan's regulatory criteria for landfilling of 0.3mg/L.

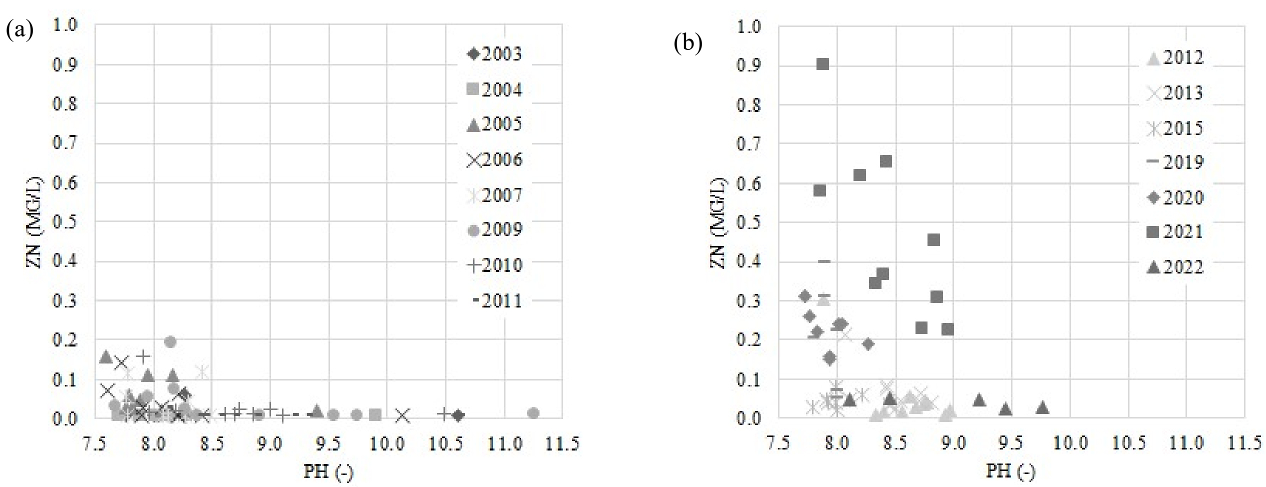

Fig 3. Zn vs pH graph with actual data from column leaching experiment (a) 2003-2011 (b) 2012 to 2022