Stabilization of lead in municipal solid waste incineration fly ash using a combined treatment of hydroxyapatite from fishbone and a chelating agent

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1.Introduction

Currently, 2.92 million tons of incineration residues of municipal wastes are generated annually in Japan.[1)] Incineration residues mainly consist of bottom ash and fly ash. Fly ash contains hazardous heavy metals such as lead and is required to undergo treatment to control the elution of heavy metals before final disposal. However, existing stabilization methods are costly and consume a large amount of landfill space. At the same time, the annual consumption of fish and shellfish in Japan is 7.16 million tons, and a large amount of fish sludge is generated.[2)] It has been known that hydroxyapatite contained in fishbones reacts with heavy metals such as lead to form insoluble phosphate as follows:[3)]

$$Ca_5(PO_4)_3(OH) + H_2O \rightarrow 5Ca^{2+}(aq) + 3PO_4{}^{3-}(aq) + 2OH^-(aq)$$
$$5Pb^{2+}(aq) + 3PO_4{}^{3-}(aq) + 2OH^-(aq) \rightarrow Pb_5(PO_4)_3(OH) + H_2O$$

Previous studies have also examined the effects of liquid-solid ratio, setting time, and fish species on lead stabilization. However, the effect of fishbone treatment alone has not been sufficient to lower lead concentration to the landfill decision criteria. Therefore, in this study, to clarify the lead stabilization process in fly ash with fishbone and a chelating agent, three treatments were carried out: only fishbone, only chelating agent, and a mixture of fishbone and chelating agent. Then, we compared their immobilization effects.

2. Meterials and methods

2-1 Materials

Fly ash samples from four waste to energy plants (N, H, R, and J) were used and hereinafter referred to as N, H, R, and J. A piperazine chelating agent was used as the chelating agent.

2-2 Methods

(1) Treatment using only fishbone

Ten grams of fly ash and 1g of fishbone were placed into a 250 ml polyethylene container, and 15ml of pure water was added to reach the liquid-solid ratio of 1.5. Then, each container was stirred at 120 rpm for 1 min. The incubation time was set at 6h, 12h, 24h, 3d, 7d, 14d, and 28d. After each settlement period, JLT-13 was conducted.

(2) Treatment using only chelating agent

Ten grams of fly ash was placed into a 250 ml polyethylene container, and 0 to 2.5% chelating agent and 3 ml of pure water were added. Each container was stirred at 120 rpm for 2 minutes, and the container was set at 1 hour with no lid. After that, JLT-13 was conducted.

(3) Combined treatment with fishbone and chelating agent

In a 250 ml polyethylene container, 10g of fly ash and 1g of fishbone were placed, and pure water was added. The liquid-solid ratio was maintained at 1.5. After that, 0 to 2.5% chelating agent was added, and the container was set at 1 hour with no lid. Finally, JLT-13 was conducted.

2-3 Results and discussion

The dashed line in Figure 1 indicates the standard value of 0.3 mg/L for Pb in the landfill. The N fly ash, which had the lowest initial Pb concentration of 3.25 mg/L, satisfied the landfill standard in 6 hours. Figure 2 shows that the initial concentrations of the two samples differed by more than a factor of two, but after 7 d, the Pb concentration in the R fly ash was lower. Elemental analysis of the fly ash was performed by XRF, considering that the reactions are different depending on the elemental composition of fly ash. Focusing on the differences in the percentage of calcium compound content, the relationship between the pH of each fly ash and the setting time is shown in Figure 3. The pH of fly ash samples H and J with high calcium content did not considerably change, while the pH of N and R fly ashes with low calcium content (<40%) showed a decreasing trend over time. This suggests that a low pH of 11.5 or lower promotes stabilization in a shorter period (less than one day).Figure 4 shows the relationship between the Pb concentration in the J fly ash and the chelating agent addition rates. The combined treatment satisfied the landfill standard at an additional rate of 1.3%, while the chelate alone satisfied the landfill standard at an additional rate of 1.5%. In all cases (except one), the Pb concentration was lower in the combined treatment than in the chelate treatment.

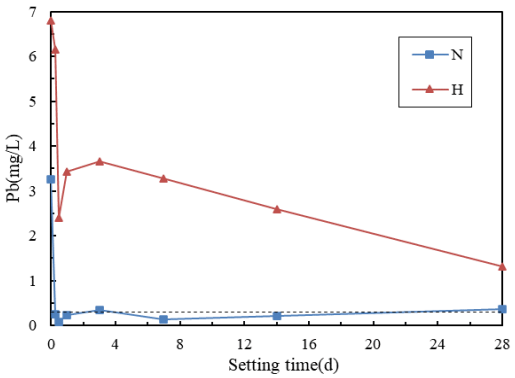

Figure 1. The relationship between Pb concentration and setting time (N and H).

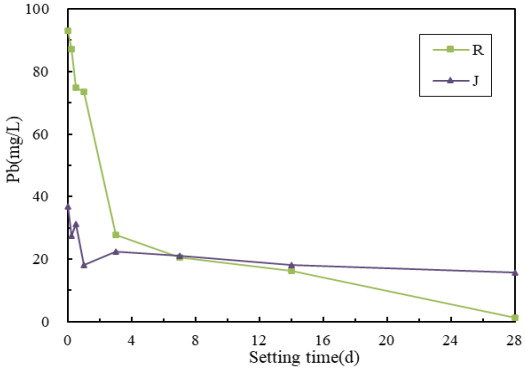

Figure 2. The relationship between Pb concentration and setting time (R and J).

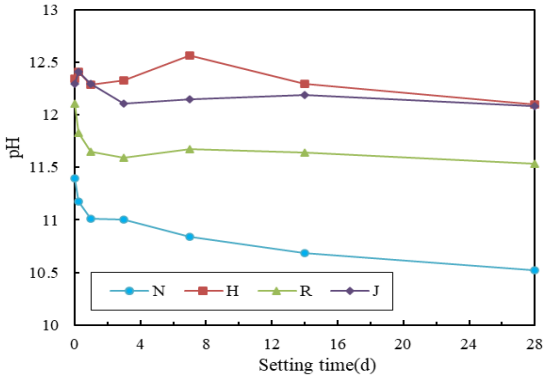

Figure 3. The relationship between pH and setting time.

3.Conclusions

In fly ashes with low Pb leaching concentrations (less than 3.25 mg/L), treatment with fishbone only could satisfy the landfill standard. In the fishbone treatment, it was found that the stabilization of lead was accelerated in shorter periods (less than 1 day), especially when the content of calcium in the fly ash was less than 40% and the pH was lower than 11.5. The combined treatment was shown to be up to 50% more effective than chelate (only) in stabilizing lead.

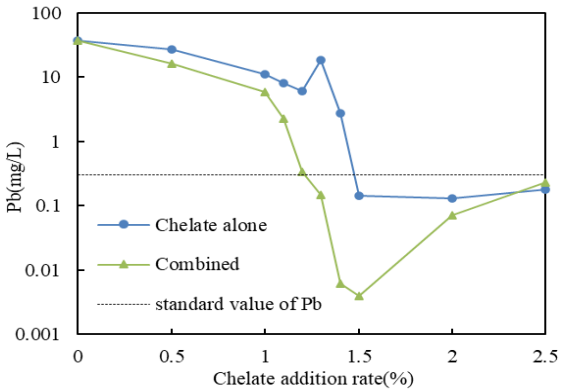

Figure 4. The relationship between Pb concentration and chelate addition rate.

[Reference]

1) Ministry of the Environment: Waste Management in Japan, Fiscal Year 2008 Edition, 2020 2) Fisheries Agency: White Paper on Fisheries in Fiscal Year 2008, 2019 3) Q. Y. Ma, S. J. Traina, T. J. Logan : In Situ Lead Immobilization by Apatite, Environ. Technol., 1993, Vol. 27, No. 9, pp. 1803-1810