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Solidification/Stabilization of Low-Level Radioactive Wastes including Hazardous Substances from Uranium Fuel Processing Plants

Junya SATO*, Yuta TAKAHASHI, Jun SUNAHARA, Toshimitsu SAITO,
Yukihiko YOSHIDA, Tomoyuki SONE and Takeshi OSUGI

Japan Atomic Energy Agency, 4-33 Muramatsu, Tokai-mura, Naka-gun, Ibaraki-ken, 319-1194, Japan

For low-level radioactive waste containing hazardous substances (mixed wastes) generated from uranium fuel processing plants, establishing appropriate solidification/stabilization methods is one of the key challenges for their safe and effective storage and disposal in Japan. This study investigated the solidification/stabilization methods of the mixed waste sludge containing hazardous substances of lead, cadmium and mercury by using various solidification materials. Additionally, the compressive strength of solidified products was investigated along with the leaching behavior of hazardous substances.

KEYWORDS: *Hazardous and Radioactive Waste, Solidification/Stabilization, Cement, Leaching*

I. Introduction

Uranium waste is generated from uranium fuel processing plants including mining and milling of uranium, conversion, enrichment and fuel fabrication, and the waste is categorized in Japan into metals, concrete rubble, miscellaneous wastes, sludge, and filters. Among these wastes, sludge is generated by treatment of uranium-contaminated liquid waste from processes such as reconversion, molding, analysis, washing, cleaning, and other related operations. As a result, the types of sludge vary depending on coagulation-sedimentation treatment processes, such as iron sludge, silica sludge, poly-aluminum chloride sludge, soil, as well as other types¹⁾. For these sludge wastes, many technical issues remain to be clarified for safe treatment and disposal, because they are generated in large amounts and have high concentrations of uranium and hazardous substances such as lead (Pb), cadmium (Cd) and mercury (Hg). While provisional limits for radionuclide concentrations in certain radioactive waste have been established, the limits for hazardous substances in radioactive waste have not yet been determined²⁾. In Japan, the concentrations of hazardous substances released from industrial wastes are regulated under the Waste Disposal and Public Cleansing Law. If similar regulations are introduced for hazardous substances in radioactive waste, it will be crucial to develop conditioning techniques that stabilize these materials while also assessing their environmental impact on surrounding disposal facilities.

Our commissioned research investigated techniques for reducing the volume of low-level radioactive sludge containing hazardous substances (mixed wastes) by recovering high-concentration uranium from the sludge. Additionally, the solidification and stabilization of uranium-recovered mixed waste was also studied in collaboration with uranium processing manufacturers. This commissioned research examined four types of mixed waste: iron sludge,

silica sludge, aluminum chloride sludge, and soil. Of these, iron sludge is one of the most abundant types of mixed waste. It is generated by adding iron chloride and an alkaline reagent to liquid waste from equipment, followed by sediment collection and drying. This sludge contains not only uranium but also hazardous heavy metals such as Pb and Hg.

This paper focuses on investigating suitable methods and conditions for solidification and stabilization of iron sludge by using the simulated sludge without radioactive nuclides. This approach was adopted to ensure that the concentrations of heavy metals in the leachate from waste forms comply with environmental regulatory limits. In general, the solubility of Pb, an amphoteric metal, in aqueous solution is expected to increase in highly alkaline conditions due to the presence of Ordinary Portland Cement (OPC), a widely used material for the solidification of radioactive waste. Additionally, there are concerns regarding the potential solubility of other heavy metals in highly alkaline conditions from the solidified products, which may vary depending on the specific type of heavy metal present^{3,4)}. To address these challenges, four candidate solidification materials were selected based on differences in their pH levels: Alkali-Activated Material (AAM), OPC, Low Alkaline Cement (LAC), and Chemically Bonded Phosphate Ceramics (CBPC).

Several previous studies have reported the immobilization of Pb, Cd, and Hg using OPC. Stabilizing tailing waste containing Pb and Cd with OPC has been shown to meet US standards⁵⁾. However, while OPC effectively stabilizes Cd, the degree of Pb stabilization is lower due to differences in pH dependency among elements^{6,7)}. Studies on AAM have also reported varying degrees of Pb and Cd immobilization, which is attributed to the variability of AAM composition, affecting its performance⁸⁻¹²⁾; CBPC has been reported to simultaneously stabilize Pb, Cd, and Hg¹³⁾. On the other hand, few studies have examined LAC, although mercury sulfide stabilization using LAC has been evaluated according to Japanese standards^{14,15)}.

Most previous studies used different leaching tests, making

*Corresponding author, E-mail: sato.junya@jaea.go.jp

it difficult to compare solidification materials under consistent conditions. Additionally, solidification test conditions vary across studies, complicating direct performance comparisons. Therefore, this study conducted leaching tests for Pb, Cd, and Hg using four solidification materials under identical conditions based on Japanese standard methods to systematically compare their performance. The maximum concentration of each heavy metal in the solidified materials was estimated to ensure compliance with Japanese standards. Nitric acid salts of Pb, Cd, and Hg were added to the solidified samples to evaluate their immobilization capacity for heavy metals. These salts were selected for their high-water solubility to enable effective assessment of the solidification materials.

For the solidification tests of simulated iron sludge, two solidification materials were selected based on their performance in stabilizing heavy metals. The maximum waste loading of the simulated iron sludge was determined based on the compressive strength standard, which ensures the structural integrity of radioactive waste forms¹⁶⁾, as well as the concentration of heavy metals in the leaching solution (leachate) to evaluate their immobilization efficiency and compliance with environmental regulations.

II. Experimental

1. Solidification/Stabilization of Hazardous Substances

OPC, LAC, AAM, and CBPC were used as solidification materials. The raw materials used for these solidification materials are listed in **Table 1**. Deionized water (DIW, 18.2 MΩ·cm) was used as the mixing water.

For the solidification and stabilization tests of hazardous substances, Pb, Cd, and Hg were identified as target heavy metals based on previous analytical data from actual sludge waste. Lead nitrate ($\text{Pb}(\text{NO}_3)_2$, 99.9%, FUJIFILM Wako Pure

Chemical Corp., Japan), cadmium nitrate tetrahydrate ($\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, >98.0%, Kanto Chemical Co., Inc., Japan), and mercury nitrate ($\text{Hg}(\text{NO}_3)_2$, 0.1 mol/L, Hayashi Pure Chemical Ind., Ltd., Japan) were used as metal sources for preparation of heavy metal-containing water.

The heavy metal-containing water was prepared by dissolving certain amounts of $\text{Pb}(\text{NO}_3)_2$, $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, and $\text{Hg}(\text{NO}_3)_2$ in DIW. Then, the raw solidification materials were added in the heavy metal-containing water to be mixed with a mechanical mixer (MM400, Retsch GmbH, Germany) for 4 minutes. After mixing, the pH of mixed pastes was measured by using a pH meter (F-74, HORIBA Ltd., Japan), and the pastes were poured into a polypropylene tube with sealing and cured at 20°C for 28 days in an incubator (ILD-120, Alp Co., Ltd., Japan) to provide the solidified products. The chemical composition of solidified products is shown in **Table 2**.

A leaching test was conducted on the cured solidified products according to Japanese Environmental Agency Notification No. 13 (1973), which is the Japanese standard procedure for industrial waste. This method is currently under consideration for application in evaluating heavy metal leaching from low-level radioactive waste. The solidified products were crushed and sieved to obtain particles ranging from 4.75 mm to 0.50 mm. A 2.5 g portion of the crushed sample was placed into 50-mL polypropylene centrifuge tubes, and 25 mL of DIW was added. The tubes were shaken for 6 hours at an amplitude of 4 cm and 200 rpm at 20°C. Following centrifugation at $3000 \times g$ for 20 minutes, the supernatant was filtered through a 1.0-μm membrane filter. The Pb and Cd concentrations in the leachate were analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, iCAP6000Duo, Thermo Fisher Scientific Inc., Germany), while Hg was measured using a mercury analyzer (HG-450, HIRANUMA Co., Ltd., Japan).

Table 1 Materials and preparation solutions used for solidification.

	OPC	LAC	AAM	CBPC
Material 1	OPC (Japan Cement Association)	LAC (Taiheiyo Consultant Co., Ltd.)	PoleStar 450 (Imerys Minerals Japan)	Calcium oxide
Material 2	-	-	-	Sodium dihydrogen phosphate
Mixing solution	DIW	DIW	DIW, Sodium silicate (WG), Sodium hydroxide (NaOH)	DIW

Table 2 Chemical composition of solidified products in the solidification/stabilization test for hazardous substances.

Sample	Material 1 [wt.%]	Material 2 [wt.%]	Mixing solution [wt.%]			Heavy metals [wt.%]		
			DIW	WG	NaOH	Pb	Cd	Hg
OPC	66.6	-	33.3	-	-	0.01	0.01	0.0001
	66.6	-	31.3	-	-	1.0	1.0	0.001
	66.6	-	28.3	-	-	2.5	2.5	0.01
LAC	66.6	-	33.3	-	-	0.01	0.01	0.0001
	66.6	-	31.3	-	-	1.0	1.0	0.001
	66.6	-	28.3	-	-	2.5	2.5	0.01
AAM	30.1	-	27.7	34.0	8.0	0.01	0.01	0.0001
	30.1	-	27.2	34.0	8.0	0.25	0.25	0.001
	30.1	-	25.7	34.0	8.0	1.0	1.0	0.01
CBPC	21.0	45.0	34.0	-	-	0.01	0.01	0.0001
	21.0	45.0	32.0	-	-	1.0	1.0	0.001
	21.0	45.0	29.0	-	-	2.5	2.5	0.01

2. Solidification/Stabilization of Simulated Sludge Wastes

The simulated iron sludge was obtained from uranium processing manufacturers. The procedure was a scaled-down adaptation of the actual process used in uranium processing facilities. Initially, 60 liters of a solution containing Pb, Cd, and Hg were prepared, with a surfactant added to replicate the characteristics of industrial wastewater. To form the sediment, 30 mL of polyferric sulfate and 50 g of barium chloride were added to the solution, and the pH was adjusted to 7 using a sodium hydroxide solution. The resulting sediment was mixed with 75 g of diatomite, then dried at 120°C for 30 hours before being used as the simulated iron sludge in the tests. The heavy metal concentrations in the sludge were intentionally set higher to account for variations in actual sludge composition. The prepared sludge mixture was designed to contain 0.81 wt.% Pb, 0.98 wt.% Cd, and 0.39 wt.% Hg, reflecting typical variations in sludge composition.

The iron sludge was solidified using OPC and LAC at loading ratios of 10, 30, and 50 wt.%. The solidified products were labeled accordingly; for example, OPC with the iron sludge loading of 10 wt.% was denoted as OPC-Fe10%, while a sample without any sludge was labeled as BL. The 10 wt.% loading sample contained 0.08 wt.% Pb, 0.10 wt.% Cd, and 0.04 wt.% Hg, whereas the 50 wt.% loading sample contained 0.41 wt.% Pb, 0.49 wt.% Cd, and 0.20 wt.% Hg. The water-to-cement ratio was maintained at 0.50, with the water content adjusted based on the water absorption capacity of the iron sludge. The mixing procedure followed the methodology described in the “Solidification/Stabilization of Hazardous Substances” section. The prepared paste was cast into 20-mm cubic molds and cured at 20°C for 28 days in an incubator. After curing, the compressive strength of the solidified products was measured using an automatic compression testing apparatus (Hi-ACTIS-200L, MARUI Co., Ltd., Japan). Additionally, a leaching test was conducted according to the procedure described in the “Solidification/Stabilization of Hazardous Substances” section.

III. Results and Discussion

1. Solidification/Stabilization of Hazardous Substances

The pH of mixed pastes varied depending on the solidification material, and it was pH 14 for AAM, pH 13 for OPC, pH 11 for LAC, and pH 6 for CBPC. This confirmed

that the pH range of mixed pastes differs based on the type of solidification material used.

Figure 1 presents the leaching test results on heavy metal concentration in leachate as a function of the concentration in the solidified products for each heavy metal. For Pb, AAM with the Pb concentration of 0.01 wt.% complies with the Japanese standard of leachate concentration ($Pb \leq 0.3$ mg/L). OPC with the Pb concentration less than 0.1 wt.% met the standard, while LAC and CBPC satisfied the standard even at the Pb concentrations as high as 2.5 wt.%. Both OPC and AAM, where a certain leaching of Pb was detected, are highly alkaline solidification materials. In contrast, LAC and CBPC exhibited no detectable Pb leaching, as they are lower in alkalinity. The OPC results aligned with those of previous studies, which reported 5 mg/L for OPC with Pb concentration of 10 wt.% and 45 mg/L for OPC with Pb concentration of 15 wt.%¹⁷⁾. However, the AAM results were higher than those in previous reports, which indicated 0.80 mg/L for AAM with Pb concentration of 3 wt.%¹⁸⁾, according to the Chinese National Standard (HJ557-2009)¹⁹⁾. Variations in leaching values likely arise from differences in test procedures among countries. Since leaching is influenced by conditions such as pH, these findings highlight the importance of standardized procedures for accurate comparisons.

For Cd, AAM exhibited the highest leaching, and only the sample with the Cd concentration of 0.01 wt.% or less met the Japanese standard of leachate concentration ($Cd \leq 0.09$ mg/L). LAC and CBPC met the standard at the Cd concentrations up to 1 wt.%, while OPC remained compliant even at the Cd concentrations of 2.5 wt.%. These results align with those of previous studies, which reported 1.29 mg/L for AAM sample with the Cd concentration of 3 wt.% and 0.817 mg/L for CBPC sample with the Cd concentration of 6 wt.%^{18,20)}. For OPC samples, Cd was not detected likely due to its high stability under highly alkaline conditions²¹⁾. However, Cd was leached from AAM despite its high alkalinity, suggesting that factors other than pH, such as the mechanical properties of the solidified products, influenced these results.

Regarding Hg, AAM exceeded the Japanese standard of leachate concentration ($Hg \leq 0.005$ mg/L) even when the Hg concentration in the sample was as low as 0.0001 wt.%. OPC and CBPC met the standard at Hg concentrations up to 0.0001 wt.%, while LAC demonstrated the highest immobilization

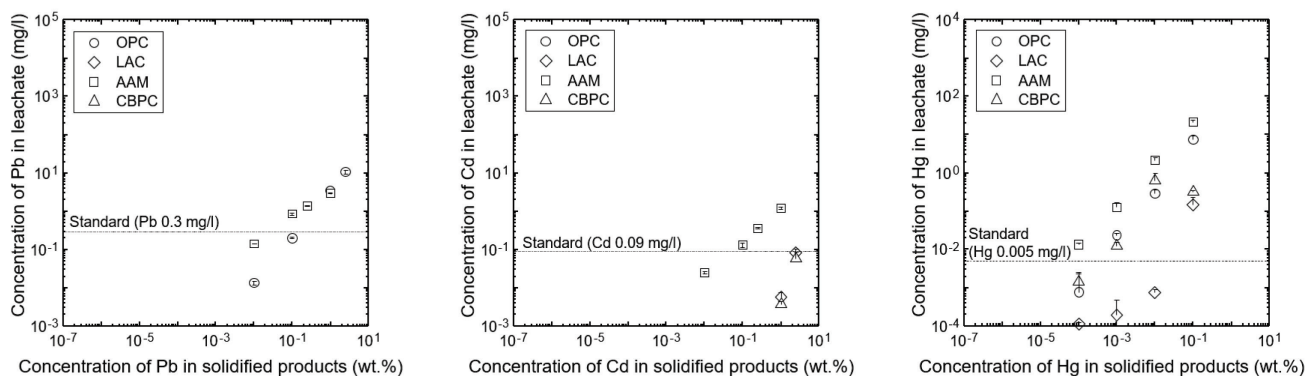


Fig. 1 Comparison between heavy metal content in the solidified products and the corresponding concentrations in the leachate. Pb leaching from LAC and CBPC was below the detection limit, while Cd leaching from OPC was not detected.

ability, remaining restrained even at 0.01 wt.%.

Pb is an amphoteric metal, and Hg compounds also exhibit chemical properties that increase leaching under highly alkaline conditions²²). Therefore, the low alkalinity of LAC contributed to greater stabilization of Pb and Hg compared to other solidification materials. Additionally, LAC is a solidification material with a composition adjusted by incorporating haunynite and belite, designed to maintain low alkalinity²³). As a result, the formation of portlandite and monosulfate hydrate, which typically induce high alkalinity in cement, is suppressed. Instead, C-S-H gel and ettringite are formed, creating a dense structure²⁴). C-S-H gel and ettringite also contribute to the stabilization of hazardous elements²⁵), which accounts for the high stabilization capacity of LAC for Pb and Hg.

Based on these findings, LAC, which demonstrated the highest immobilization of Pb and Hg, and OPC, which exhibited the highest immobilization efficiency for Cd, were selected as the solidification materials for the solidification test of simulated sludge waste.

2. Solidification/Stabilization of Simulated Sludge Wastes

Figure 2 shows the external appearance of the iron sludge solidified products prepared with OPC or LAC. Samples with the sludge loading of 10 wt.% and 30 wt.% successfully hardened after 28 days of curing, whereas those with the sludge loading of 50 wt.% were extremely brittle, and some could not be demolded.

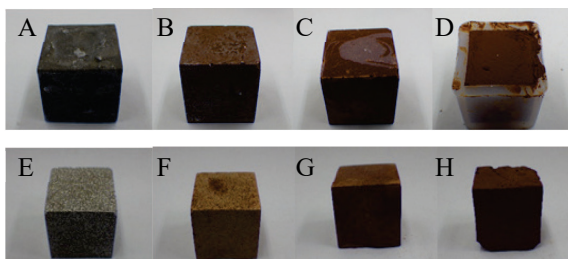


Fig. 2 Solidified OPC and LAC products containing simulated iron sludge. (A) OPC-BL, (B) OPC-Fe10%, (C) OPC-Fe30%, (D) OPC-Fe50%, (E) LAC-BL, (F) LAC-Fe10%, (G) LAC-Fe30%, and (H) LAC-Fe50%

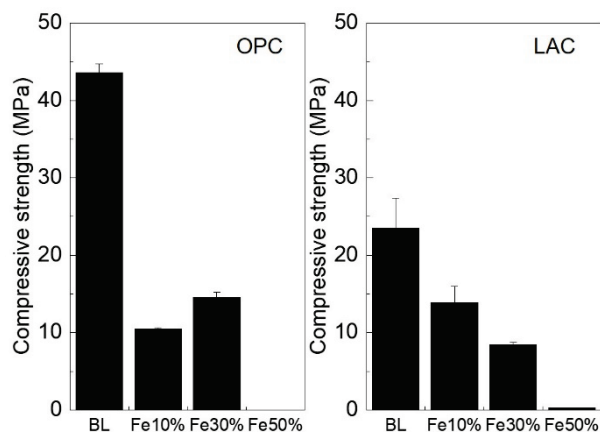


Fig. 3 Compressive strength of OPC and LAC after 28 days of curing as a function of iron sludge loading

Figure 3 illustrates the compressive strength of the solidified products. The compressive strength of LAC decreased as the iron sludge loading ratio increased, whereas OPC-Fe30% exhibited a slight increase in strength compared to OPC-Fe10%. The compressive strength met the standard value of 1.47 MPa for loading ratios up to 30 wt.% in both OPC and LAC.

The crystalline phases of the solidified products analyzed by using XRD are shown in **Fig. 4**. Portlandite was detected as the primary structural phase in OPC solidified products, whereas ettringite was the dominant phase in LAC solidified products. As the loading ratio of simulated iron sludge increased, barium sulfate was detected in both OPC and LAC samples, as expected, since it is a byproduct of the simulated iron sludge. In OPC-Fe50% and LAC-Fe50%, the main structural phase disappeared, and barium sulfate became the dominant peak, suggesting that solidification was incomplete.

Figure 5 shows the pore size distribution of the solidified products. As the iron sludge content increased, the number of large pores also increased. A previous study classified pores into four categories based on size: gel pores (less than 10 nm), transitional pores (10 to 100 nm), capillary pores (100 to 1000

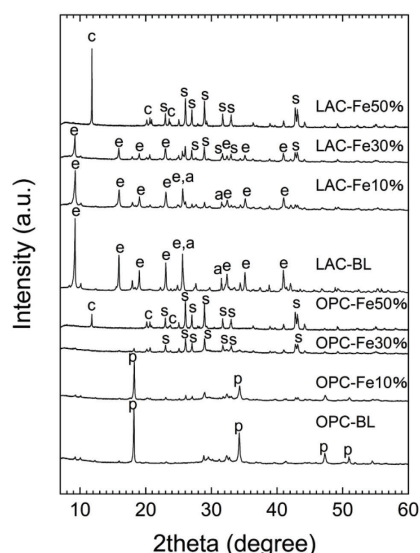


Fig. 4 X-ray diffraction patterns of OPC and LAC samples after 28 days of curing, showing portlandite (p), ettringite (e), anhydrite (a), calcium sulfate (c), and barium sulfate (s)

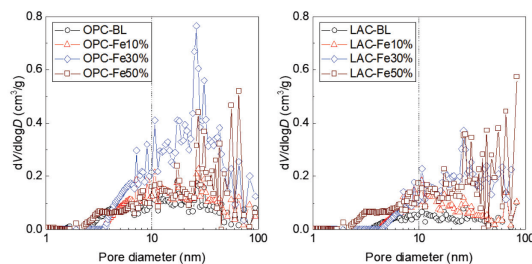


Fig. 5 Pore size distribution of OPC and LAC solidified products after 28 days of curing

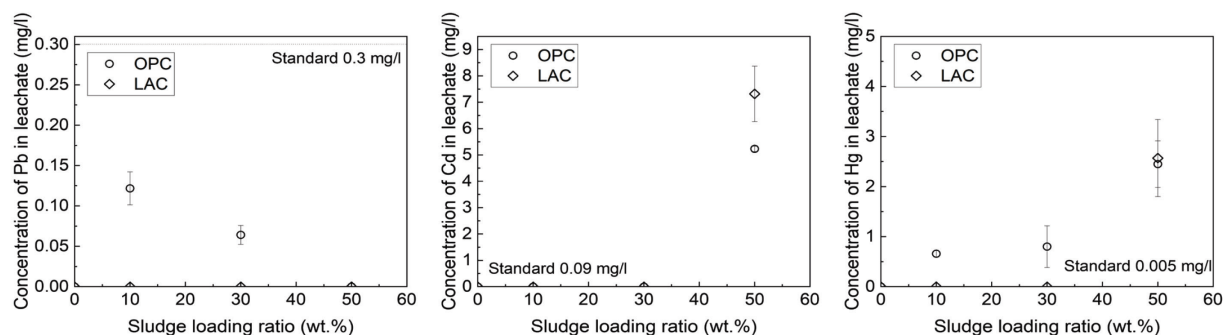


Fig. 6 Relationship between heavy metal concentration in the leachate and loading ratio of simulated iron sludge

nm), and macropores (greater than 1000 nm). While gel pores have no impact on compressive strength, an increase in capillary pores and macropores negatively affects mechanical strength²⁶⁾. The reduction in compressive strength observed with increasing iron sludge content was attributed to the formation of larger pores. Moreover, the pore distribution in OPC-Fe50% was nearly identical to that of LAC-Fe50%, suggesting that hardening was incomplete in both samples. Additionally, OPC-Fe30% exhibited an increase in gel pores compared to OPC-Fe10%, indicating a greater presence of amorphous phases, which likely contributed to the slight increase in compressive strength. Furthermore, LAC exhibited lower porosity than OPC, reinforcing its denser microstructure and lower alkalinity, which contributed to the reduced leaching of heavy metals.

Figure 6 illustrates the leaching test results on heavy metal concentration in leachate as a function of iron sludge loading ratio. The leaching of Pb met the Japanese standard of leachate concentration ($Pb \leq 0.3$ mg/L) in all cases. However, Pb concentration in leachate from OPC-Fe10% was higher than that from OPC-Fe30%, even though the total Pb content in OPC-Fe10% was lower than that in OPC-Fe30%. The pH of the leachate decreased as the proportion of OPC in the solidified product decreased, with OPC-Fe10% recording a pH of 13.4, while OPC-Fe30% had a pH of 13.1. These results suggest that lower pH levels in the leachate suppressed Pb leaching. For Cd, the leaching met the Japanese standard of leachate concentration ($Cd \leq 0.09$ mg/L) for loading ratios up to 30 wt.% in both OPC and LAC. In contrast, Hg leaching from OPC exceeded the Japanese standard of leachate concentration ($Hg \leq 0.005$ mg/L), whereas that from LAC remained below the standard except at 50 wt.% sludge loading. These results indicate that LAC solidified products containing up to 30 wt.% iron sludge complied with the heavy metal leaching standards.

The leaching results for solidified products with the sludge loading of 10 wt.% were consistent with those obtained using chemical reagents, as shown in Figure 1. However, leaching from solidified products with 30 wt.% iron sludge was lower than that shown in Figure 1, likely due to a pH reduction in the leachate, which suppressed heavy metal leaching. Specifically, the pH of OPC-Fe10% leachate was 13.4, while that of OPC-Fe30% was 13.1, supporting the observation that lower pH reduces Pb and Hg leaching^{3,22)}.

Based on these findings, the maximum loading ratio that

satisfies both strength and leaching standards was determined. In terms of compressive strength, both OPC and LAC met the required standard for loading ratios up to 30 wt.%. However, for leaching performance, OPC exceeded the standard for all samples, indicating that Hg stabilization was challenging for OPC. In contrast, LAC met the leaching standard up to a 30 wt.% loading ratio. In summary, LAC proved to be more effective in reducing heavy metal leaching, particularly when leachate concentrations exceeded the regulatory limits in OPC-based solidified products.

IV. Conclusions

This study investigated the solidification of hazardous substances and non-radioactive simulated sludge using various solidification materials with different pH levels. In the hazardous substances test, the maximum concentration of hazardous elements that satisfied the Japanese standard was identified for each solidification material. In the solidification test of simulated iron sludge, the compressive strength of the solidified products met the Japanese standard when the filling ratio was up to 30 wt.% for both OPC and LAC. The leaching test results indicated that the concentrations of Pb and Cd were effectively suppressed in the solidified products, remaining within the limits of the Japanese standard. However, Hg leaching from OPC exceeded the regulatory limit.

These findings suggest that LAC is more suitable for stabilizing Hg. When OPC is used as a solidification material, it is essential to consider the Hg concentration in the waste.

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