Nuclear Engineering and Technology 55 (2023) 2889–2896

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Leachability of lead, cadmium, and antimony in cement solidified waste in a silo-type radioactive waste disposal facility environment

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ARTICLE INFO

Article history:
Received 17 January 2023
Received in revised form
8 April 2023
Accepted 8 May 2023
Available online 9 May 2023

Keywords:
Cementitious waste
Heavy metal
Leaching test
Mixed waste
TCLP

ABSTRACT

The waste acceptance criteria for heavy metals in mixed waste should be developed by reflecting the leaching behaviors that could highly depend on the repository design and environment surrounding the waste. The current standards widely used to evaluate the leaching characteristics of heavy metals would not be appropriate for the silo-type repository since they are developed for landfills, which are more common than a silo-type repository. This research aimed to explore the leaching behaviors of cementitious waste with Pb, Cd, and Sb metallic and oxide powders in an environment simulating a silo-type radioactive waste repository. The Toxicity Characteristic Leaching Procedure (TCLP) and the ANS 16.1 standard were employed with standard and two modified solutions: concrete-saturated deionized and underground water. The compositions and elemental distribution of leachates and specimens were analyzed using an inductively coupled plasma optical emission spectrometer (ICP-OES) and energy-dispersive X-ray spectroscopy combined with scanning electron microscopy (SEM-EDS). Lead and antimony demonstrated high leaching levels in the modified leaching solutions, while cadmium exhibited minimal leaching behavior and remained mainly within the cement matrix. The results emphasize the significance of understanding heavy metals' leaching behavior in the repository's geochemical environment, which could accelerate or mitigate the reaction.

1. Introduction

The management of mixed wastes, which contain both radioactive and non-radioactive hazardous materials, poses considerable challenges due to the risks associated with both types of materials. In nuclear facilities, mixed wastes containing heavy metals, asbestos, and organic solvents are continuously generated from activities such as radiochemical experiments, decommissioning, and radioisotope analysis [1]. To properly dispose of mixed wastes, waste acceptance criteria (WAC) must be established by carefully considering the geochemical behavior of the radioactive isotopes and hazardous materials, as well as their interaction with the wasteform and leaching solution [2,3]. However, establishing WAC for mixed waste is complicated by the complex behavior of mixed wastes in different repository sites, which makes it difficult to develop standard criteria for mixed waste. As a result, only a few countries, such as the United States (U.S.), have established regulatory systems to address mixed waste management [4].

In the U.S., there are a few mixed waste disposal facilities including Hanford, which accepts waste streams for final disposal in Low-Level Burial Ground (LLBG) Trenches. Compliance with the applicable Land Disposal Restriction (LDR) requirements, as stated in 40 CFR 268.7 (b), is mandatory prior to disposal, and this is assessed by the Toxicity Characteristic Leaching Procedure (TCLP). However, studies have shown that TCLP may not be a reliable indicator of the environmental conditions of trench-type disposal sites due to differences in pH and composition between the TCLP leaching solution (typically acetic acid with pH 2.88 or 4.93) and the actual leachate in the repository [5,6]. In a silo-type repository usually constructed with concretes and situated several hundred meters underground, these differences are expected to be even greater. The leachate in the repository will be highly alkaline due to the reaction of the concrete and the presence of various ions in the groundwater [7–12].

In recent years, numerous studies have investigated the leaching behavior of toxic substances, such as heavy metals in soil [13,14], and ashes [15–17] in hazardous waste under different pH and solution composition conditions. The results have shown that the leaching behavior of metals can vary significantly depending on

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https://doi.org/10.1016/j.net.2023.05.012

the alkalinity and various groundwater constituents, resulting in different leachability according to the nature of the metal [14,18]. For instance, research on red mud indicated that lead was not leached out under a pH range of 5.5–8 but dissolved faster as pH increased. Despite these findings, research on the leaching behavior of heavy metals in cementitious waste, which is the most widely accepted wasteform for radioactive waste, remains limited in the environmental conditions of a silo-type repository.

This study explores the leaching behaviors of heavy metals (Pb, Cd, and Sb) and their oxides (PbO and Sb_2O_3) in a cementitious waste by performing two standard leaching tests, TCLP and ANS 16.1 with standard leachant and modified solutions for simulating a silo-type repository environment. The solutions mimicking the underground repository condition were prepared by saturating underground water collected near the repository in Gyeongju with concretes. Also, the distilled water saturated with concretes was used to differentiate the effects of the pH and underground water composition on leaching results. The leachate characteristics, such as acidity, electrochemical potential, and composition, were investigated by pH-Eh meters and Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) before and after the test. Heavy metal distribution on the specimen after the test were examined using energy-dispersive X-ray spectroscopy combined with scanning electron microscopy. The results would help pave the way to establish the site-specific WAC of the mixed waste for silo-type radioactive waste repositories.

2. Methods

2.1. Selection of hazardous substances

The non-radiological hazardous substances considered in low-and-intermediate radioactive waste disposal in Sweden, France, the U.K., the U.S., Canada, and Norway are represented in Table 1 [19]. Heavy metals such as Cd, Be, Pb, Cr, and Hg are used in control rods, neutron reflectors, and shielding materials and are commonly identified as hazardous in many countries. Depending on the country, other materials, such as asbestos, cyanide, and organic solvents, may also be considered hazardous [20]. Norway has selected toxic substances by the following criteria: 1) significant quantity in the predicted waste, 2) release rate from the waste, 3) transportability in the environment near the repository, 4) bioaccumulation, 5) understanding of toxic effect mechanism to humans and biota. From this perspective, antimony could be potentially listed as toxic in a silo-type radioactive waste repository for the following reasons: radioactive uranium-antimony oxides (USb_3O_{10}) has utilized as catalysts in some petrochemical industries, and the leaching of antimony would be accelerated in a highly alkaline solution of the repository. In this study, for this reason, Pb and Cd, the most commonly encountered heavy metals in nuclear facilities, and Sb, whose leachability would be raised in a silo-type repository, were selected for the leaching tests. Both the metallic form (Pb, Cd, and Sb) and oxides (PbO and Sb_2O_3) of the testing materials were utilized in the leaching test.

Table 1
List of significant hazardous substances for radioactive waste disposal.

	Substances of interest
Sweden	L/ILW - Pb, asbestos, epoxy resins ILW – organic material, Cd, Be, Pb, Cr, Hg
France	Pb, B, Ni, Cr, As, Sb, Se, Cd, Hg, Be, cyanide, U, asbestos
UK	Be, Cd, Cr, Pb, U
US	Mo, Ni, V
Canada	Hg, Pb, Cd, Cu, Cr, Ni, Mn, Pb
Norway	As, Be, Cd, Cr, Hg, Pb, U, asbestos

2.2. Preparation of waste specimens

The leaching test specimens were prepared by solidifying heavy metal metallic and oxide powders with cement, which is one of the most common ways to immobilize radioactive waste and toxicants with high stability in a repository [21,22]. The metallic (Pb, Cd, and Sb) and oxides powders (PbO and Sb_2O_3) with a size of 100 mesh supplied by Sigma Aldrich were utilized. The cement paste was prepared following the standard mixing procedure of KSL ISO 679 with 450 g of Type I Portland cement (supplied by Hanil Corporation), 200 ml of water, 120 g of metal or oxide powders, and 1.3 kg of ISO standard sand [23]. A water-to-cement (w/c) ratio and waste loading were 0.44 and 5.7 wt%, respectively. The mixed paste was poured and cured for 28 days in a cylindrical mold of 5 cm (width) × 10 cm (height) at room temperature (Fig. 1). The mold size is designed for ANS 16.1 test, and the cured cement was crushed and sieved through a 9-mm mesh to produce specimens for the TCLP test.

2.3. Leaching test

Two standard leaching tests were adopted to evaluate the leaching behavior the heavy metal in the simulated environment of a silo-type repository. Given the potential danger associated with mixed waste, which may include both chemical hazardousness and radioactivity, TCLP, which is widely used for the evaluation of toxicity of municipal wastes, and ANS 16.1, a leaching test method specifically designed for assessing the leaching property of radioactive wastes were utilized.

2.3.1. TCLP [24]

TCLP is a standardized leaching test method developed by the United States Environmental Protection Agency (USEPA) to examine the characteristic toxicity in a municipal solid landfill facility. The TCLP is an approved method under the Federal Resource Conservation and Recovery Act (RCRA) hazardous waste regulations, thereby widely utilized in environmental and waste management studies. For non-volatile solid samples, a minimum of 100 g waste sample is required. The waste sample should be thoroughly dried, crushed, and sieved so that the largest side is smaller than 9.8 mm. The leaching solution is determined via preliminary evaluation figuring out the acidity characteristic of the waste. In this study, the leachant of extraction #1 was selected, which was prepared by dissolving glacial acetic acid (100%, supplied by Sigma Aldrich) in deionized water, followed by adjusting the pH to 4.93 ± 0.05 using HCl (37%, supplied by Sigma Aldrich) or NaOH (1 M, supplied by Sigma Aldrich) solutions. Following the procedure, the weight of the extraction fluid was 20 times of the

Fig. 1. Waste samples produced with cement solidification.

waste sample. The jar containing waste samples and leachant was rotated 360° at 30 ± 2 rpm for 18 ± 2 h using a rotary agitator. Then, the solution was filtered by a borosilicate glass fiber filter and compositional changes were analyzed by ICP-OES.

After the TCLP test, the surface of the waste sample that was filtered from the leaching fluid was analyzed using a scanning electron microscope and an X-ray spectrometer (Hitachi High-Tech Corporation, SU7000) to confirm the distribution of each metal component on the surface of the waste sample. Microscopic images were obtained at 500 × magnification with a 20 kV voltage and a working distance of 6 mm.

2.3.2. ANS 16.1

The ANS 16.1 standard test was designed by the Nuclear Regulatory Commission (NRC) to compare the leaching resistance of solidified radioactive waste [25]. The monolithic sample that accurately represents the total waste stream is recommended and should not resize the specimen size. The leaching solution, as per ANS 16.1 is deionized water at an appropriate level, as defined by ASTM D1193 Type I, II, III. The solution volume is determined by the waste sample surface area and falls within the range specified by Equation (1).

$$\frac{V}{SA} = 10 \pm 0.2 \text{ cm} \tag{1}$$

Where V is the volume of the leaching solution, and SA is the surface area of the waste sample.The specimen is immersed into the leachate by maximizing the contact area to the leachant, then leaving it for 24 ± 0.5 h at room temperature. After that, the solution is replaced with a fresh solution, waiting another equivalent interval. Repeating this procedure at least four times, the leachate is visually inspected to determine whether particles or parts of the specimen are detached. Subsequently, the solution is filtered to measure the concentration of the target element. The leached element concentration and the leaching-reaction time released through the kth leaching interval for nuclide i were calculated and presented as $y_{i,k}$, $x_{i,k}$, respectively, as shown in Equations (2) and (3):

$$y_{i,k} = (CF_{i,k})\left(\frac{V_{specimen}}{SA}\right) \tag{2}$$

$$x_i = \sqrt{\sum_{j=1}^{k} t_j} \tag{3}$$

where *CF(i,k)* is the ratio of nuclide i leaching during the leaching period k times, $\sum_{j=1}^{k} t_j$ is the cumulative leaching time during the kth leaching period, $V_{specimen}$ is the volume of the specimen, and SA is the surface area of the specimen.

The calculated data were plotted as $y_{i,k}$ versus $x_{i,k}$ for each radionuclide, and fitted using the generic linear equation (Equation (4)). The slope of the curve was then derived by linear regression in Equation (5):

$$y_{i,k} = m_i x_{i,k} + b_i \tag{4}$$

$$m_i = \frac{\sum_k x_k y_k - \left[\left(\sum_k x_k\right)\left(\sum_k y_k\right)\right]/n}{\sum_k (x_k)^2 - \left(\sum_k x_k\right)^2/n} = \frac{\sum_k (x_k - \overline{x})(y_k - \overline{y})}{\sum_k (x_k - \overline{x})^2} \tag{5}$$

where m_i is the slope derived by linear regression, b_i is the y-intercept derived by linear regression, $\overline{y}$ and $\overline{x}$ are the mean values for the data set.

Finally, the effective diffusivity (D_e) was calculated using the slope of the linear regression (Equation (6)), and the leachability index (L_i) was calculated taking logarithm to the reciprocal of the effective diffusivity in Equation (7):

$$D_e = \frac{\pi}{4} m_i^2 \tag{6}$$

$$L_i = log\left(\frac{\beta}{D_{e,i}}\right) \tag{7}$$

2.4. Leachant modification

The leaching pattern of hazardous substances in waste is greatly influenced by environmental factors such as the composition of the leaching fluid and waste, pH, and oxidation-reduction level [5,14,26]. TCLP was designed for a municipal waste landfill facility; its leachant is an acetate buffer solution at pH 4.92. Moreover, most of the leaching tests designed for the hazardous waste classification use deionized water or weakly acidic solutions (e.g., SW-846 Method 1310B, 1312, 1315, EN 12457, etc.) [13]. Contrary, silo-type radioactive waste disposal facilities are typically built with massive concrete structures [10–12]. The leaching environment in those repositories is determined by the interaction between groundwater and concrete, whose primary reaction is the dissolution of lime ($Ca(OH)_2$) in concrete into groundwater. The leachate is expected to exhibit strong alkalinity of pH 12–13 with various ions, such as Na^+, K^+, and CO_3^{2-} [27]. Owing to these differences between disposal facilities, a substantial difference would be observed in the leaching behavior of hazardous substances.

This study utilized three distinct leachant to discern the influence of pH and underground water ion constituents: standard solutions for each test and deionized water saturated by concrete and underground water saturated by concrete. For the ANS 16.1 test, deionized water was applied as the standard solution (labeled as SA), while for the TCLP test, an acetic acid solution (labeled as ST) was used. The specific composition and characteristics of the solutions is shown in Table 2. The concrete-saturated deionized water (labeled as D) was prepared by mixing 1 kg of concrete with 2 L of distilled water for three days. After the saturation, the pH was adjusted to 12.5 with the NaOH solution. The concrete-saturated groundwater (labeled as G) was prepared by mixing the groundwater collected from the areas near the Gyeongju radioactive waste repository with concrete in the same way as the concrete-saturated deionized water.

After the leaching test, the leachate was stored at 4 °C and pretreated with nitric acid, and the concentration of waste components was analyzed using ICP-OES. The procedures for groundwater collection, pretreatment, pH measurement, and metal concentration measurement were carried out according to the guidelines set forth by the Korean Waste Control Act [7].

Table 2
Chemical composition of collected groundwater and leaching solutions.

Leaching Solution	pH	Eh (mV)	Ca^{2+} (mg/L)	K^+ (mg/L)	Na^+ (mg/L)
Groundwater	7.5	−34.7	223.0	266	68.2
G	12.5	−294.5	33.0	4.4	20.5
D	12.5	−295.0	394.0	13.7	4.5
ST	4.9	108.4	<0.1	<0.1	35.5
SA	6.1	352.1	<0.1	<0.1	<0.1

Table 3
TCLP test results of waste samples using stand solution (ST) and modified solutions (D, G).

Sample	Leaching Solution	Before Leaching		After Leaching		Leaching (mg/l)	TCLP Standard (mg/l)	UTS Limit (mg/l)
		pH	Eh (mV)	pH	Eh (mV)			
Pb	ST	4.93	−31.5	11.92	−299.3	40	5.0	0.69
	D	12.5	−319	12.57	−337.0	237		
	G	12.5	−319	12.39	−326.8	593		
PbO	ST	4.93	−31.5	11.76	−290.6	6.2		
	D	12.5	−319	12.42	−328.3	190		
	G	12.5	−319	12.32	−323.0	153		
Cd	ST	4.93	−31.5	11.87	−298.2	<0.01	1.0	0.69
	D	12.5	−319	12.35	−328.4	<0.01		
	G	12.5	−319	12.42	−332.8	<0.01		
Sb	ST	4.93	−31.5	12.05	−315.2	0.87	-[a]	1.9
	D	12.5	−319	12.32	−320.1	2.41		
	G	12.5	−319	12.59	−337.3	2.86		
Sb_2O_3	ST	4.93	−31.5	11.89	−306.2	7.29		
	D	12.5	−319	12.56	−335.9	9.32		
	G	12.5	−319	12.59	−336.2	17.7		

[a] Not stated.

3. Results and discussion

3.1. TCLP

Table 3 presents the TCLP test results for Pb, Cd, and Sb samples with solution S (pH 4.93, acetic acid buffer solution), solution D (pH 12.5, concrete-saturated distilled water), and solution G (pH 12.5, concrete-saturated groundwater) employed as the TCLP leachate. The concentrations of Pb were found to be the highest among all the metals, whereas Cd exhibited very low leaching behavior below the detection limit in all solutions. If a TCLP concentration of waste exceeds the RCRA maximum concentration criteria for Pb and Cd stated in 40 CFR Part 261 Subtitle C, the waste is considered as hazardous waste [28]. While Sb is not included in the RCRA criteria, TCLP concentration of Sb containing waste can be compared with the Universal Treatment Standards (UTS) in 40 CFR Part 268 Subtitle D, which classify the hazardous constituents along with the non-wastewater and wastewater treatment standard levels [29].

The TCLP test results for the Pb and PbO samples with solution ST significantly exceeded the RCRA criteria for Pb (5.0 mg/L), indicating that both samples should be classified as hazardous waste. In the solution D and G, concentration of Pb increased almost 6 times and 15 times, respectively, while PbO sample also showed 30 times and 25 times. The pH of the solution ST after leaching was 11.92, while the pH of the modified solutions was considerably higher at 12.57 and 12.39, respectively. Results of a number of studies have demonstrated that Pb generally exhibits amorphous solubility, resulting in higher leachability in strong alkali solutions as pH rises [5]. Therefore, the leaching reaction of Pb might be activated in solution D and G, which exhibit greater alkalinity. Furthermore, the composition of ions in the leachate was found to be important in the Pb sample, as increased concentration of Pb was observed in solution G compared to solution D, but the opposite result was observed for the PbO sample.

The Sb concentration with solution ST exceeded the UTS criteria (1.19 mg/l) in Sb_2O_3 sample but did not exceeded in Sb sample, indicating that the TCLP elutes of Sb sample could be treated as non-wastewater. However, in the solution G, Sb concentrations of Sb and Sb_2O_3 increased up to 330% and 242%, respectively, exceeding the UTS value, to be classified as wastewater source. Previous studies have shown that Sb predominantly exists in the solid form of $Sb(OH)_3$ over most of the pH ranges, and converts into $Sb(OH)_4^-$ ions at pH values greater than 12. After the TCLP test, the solution ST exhibited a pH of 11.76, where Sb primarily exists in solid form. Meanwhile, solutions D and G showed pH values of 12.42 and 12.32 respectively, where Sb begins to convert into its ionic form. This result also implies that Sb has amorphous solubility similar to Pb, and its leachability increases in the highly alkaline solution. The effect of ion composition was positive in both Sb-containing samples, particularly for the Sb_2O_3 sample which showed almost doubled concentration in solution G compared to solution D.

The TCLP test conducted on the Cd sample demonstrated undetectable level of Cd in the standard and modified solutions. Generally, Cd is known as cationic metal and its leachability decreases as solution's pH rises, and show minimal leaching in alkali

Table 4
ANS 16.1 test results of waste samples using standard solution (SA) and modified solution (D).

Sample	Leaching Solution	Before Leaching		After Leaching		Leaching Index	NRC standard[a]
		pH	Eh (mV)	pH	Eh (mV)		
Pb	SA	7	−31.5	11.45	−280.5	9.03	6
	D	12.5	−319	12.58	−339.8	8.26	
PbO	SA	7	−31.5	11.43	−278.8	9.05	
	D	12.5	−319	12.23	−317.1	7.60	
Cd	SA	7	−31.5	11.42	−256	-[b]	
	D	12.5	−319	12.23	−319.1	-[b]	
Sb	SA	7	−31.5	11.71	−295.3	9.64	
	D	12.5	−319	12.54	−345.8	9.78	
Sb_2O_3	SA	7	−31.5	11.30	−270.0	9.03	
	D	12.5	−319	12.58	−339.8	8.26	

[a] For Co, Sr, Cs.
[b] Unable to calculate due to the undetectable leaching concentration.

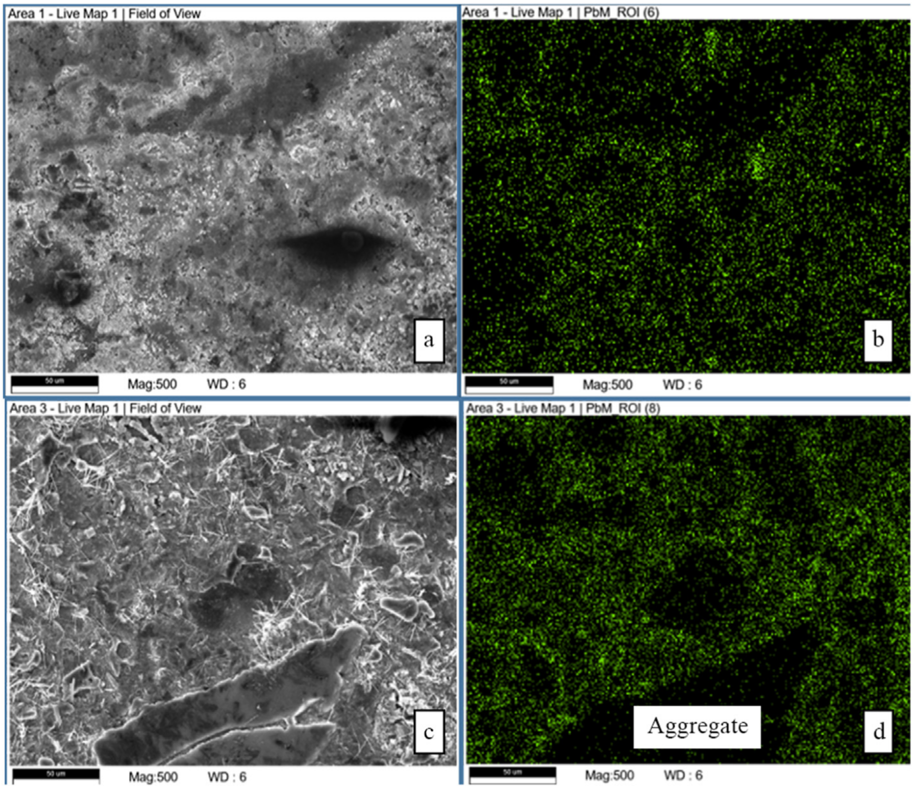

Fig. 2. Surface images of the Pb sample before TCLP (a) SEM image (b) EDS image, after TCLP (c) SEM image (d) EDS image.

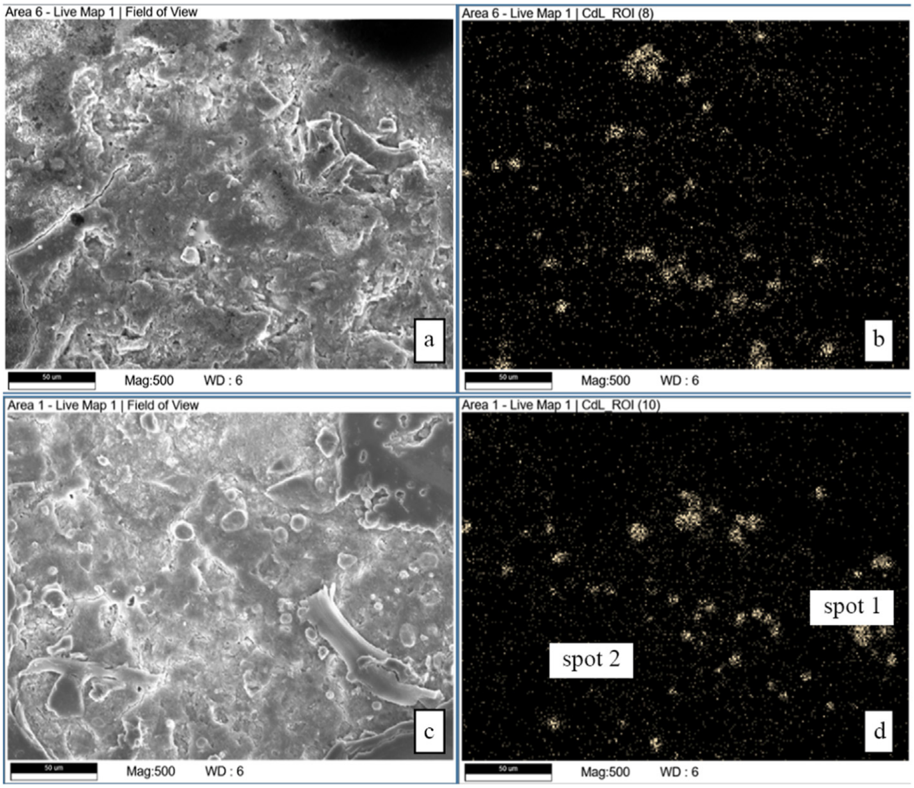

Fig. 3. Surface images of the Cd sample before TCLP (a) SEM image (b) EDS image, after TCLP (c) SEM image (d) EDS image.

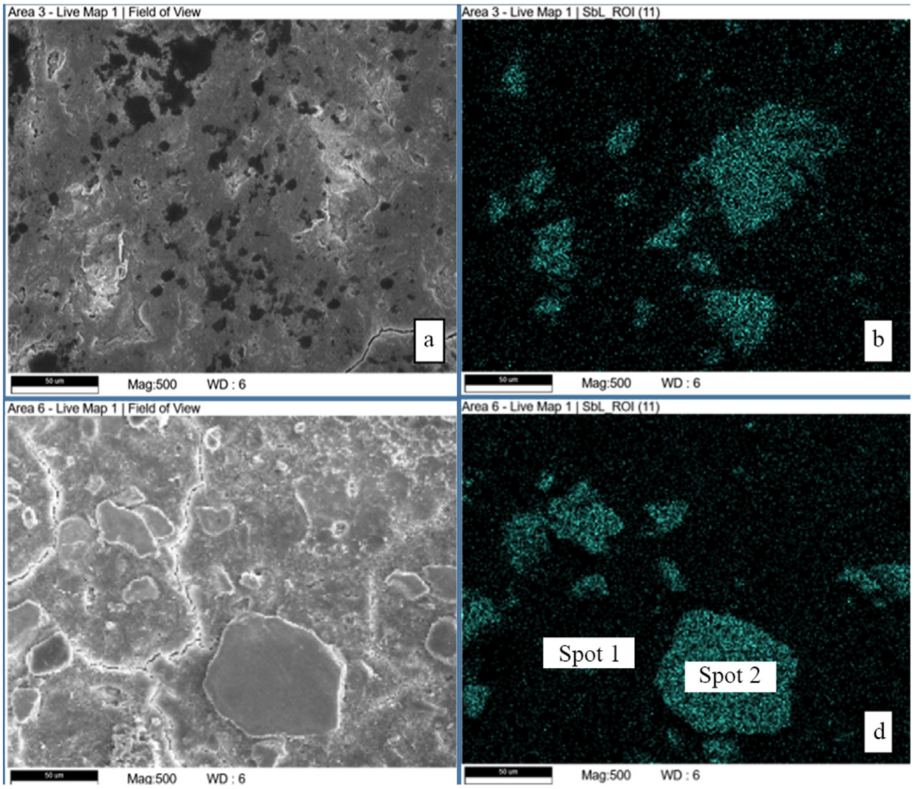

Fig. 4. Surface images of the Sb sample before TCLP (a) SEM image (b) EDS image, after TCLP (c) SEM image (d) EDS image

Table 5
EDS point analysis result of the Cd sample surface after TCLP.

Element	Weight %	
	EDS spot 1	EDS spot 2
C K	0.0	12.9
O K	17.0	47.3
Si K	0.7	7.2
Ca K	0.0	30.5
Cd L	82.3	2.0

Table 6
EDS point analysis result of the Sb sample surface after TCLP.

Element	Weight %	
	EDS spot 1	EDS spot 2
C K	2.0	12.8
O K	2.4	46.7
Si K	0.3	5.7
Ca K	2.8	17.1
Cd L	85.9	10.3

solutions with pH values higher than 6 [13]. This is due to Cd being expected to form stable solid phases minerals, such as CdS, $CdCO_3$, and $Cd(OH)_2$ [30]. In the leaching experiment, due to the abundance of ions such as CO_3^{2-} and OH^- supplied by cementitious substances in the waste sample, Cd was expected to exist in the form of stable minerals and did not leached, as the pH value of leaching solutions were much higher than 6.

3.2. ANS 16.1

For ANS 16.1 test, solution SA (distilled water) and solution D were applied as the leaching solution, and the test result is presented in Table 4. With the leaching concentration, leaching index was calculated for each of the waste samples except for the Cd sample, as the Cd concentration was undetectable. The leaching index criteria was not developed yet for the hazardous substances as the purpose of ANS 16.1 test is expressing the relative leachability of radionuclides, which is set as 6.0 only for Co, Cs, Sr to determine the availability of the disposal in the radioactive waste repository.

The test results for the Pb, PbO, Sb, and Sb_2O_3 samples indicated that the final pH of the leaching solution was higher in solution D, thus the leaching reaction occurred more actively, similar to the result of TCLP. Therefore, the leaching index, which was calculated with inverse of effective diffusivity, was higher in the solution SA for all kinds of waste samples. For the Pb sample, the diffusion coefficient gap between in solution D and SA was approximately 81 times, and in PbO sample, it was 28 times. Sb sample and Sb_2O_3 sample also showed approximately 14 times and 6 times, respectively. These findings align with the results obtained from the TCLP test, which also demonstrated the susceptibility of Pb and Sb to leaching under certain environmental conditions.

Based on the results obtained from the TCLP and ANS 16.1 test using modified leaching solution similar to the leaching environment of repository, it can be inferred that the potential leaching hazard could be more significant in a silo-type repository for the mixed wastes containing Pb, Sb, and other metal, which has a similar leaching characteristic with Pb and Sb. The effect of ion composition of leachate seems to have positive effect on increase of leachability, however, it should be supplemented for its cause and mechanisms. Moreover, these findings highlight the importance of understanding the leaching behavior of heavy metals in cement-solidified mixed waste, particularly in the context of disposal in silo-type facilities. As the leaching properties of Pb, Cd, and Sb differ significantly under varying pH conditions, careful consideration must be given to selecting appropriate treatment methods and disposal environments to minimize potential risks.

3.3. Microscopic observation

Figs. 2–4 shows the surface images of the waste samples after the TCLP test, magnified 500 times. The surfaces of the samples were analyzed to determine the distribution of metal components within the waste surface. Fig. 2(a) and (c) show microimages of the Pb sample before and after the TCLP test. The black areas at the bottom of Fig. 2(c) represents the grains of standard sand added during the mixing stage. Due to the small Pb particle size, it could not be observed in its exact particle size. Moreover, Pb was uniformly distributed in the solid solution without agglomeration. Fig. 3 and Table 5 present the surface images and EDS results of the Cd sample before and after the TCLP test. Spot 1 and 2 were analyzed to the Cd content and compared. Spot 1 was estimated to be Cd-containing substances, exhibiting 82.3% of Cd, while spot 2 showed only 2% of Cd, suggesting that the Cd powder is not dissolved within the cement matrix. In the surface image of the Sb sample waste (Fig. 4), some particles were agglomerated and presumed to contain Sb. Point analysis of spots 1 and 2 (Table 6), where Sb particles were estimated to be present, revealed approximately 85.9% and 10.3% of the Sb components, respectively.

Through the microscopic analysis of the surface image of the waste sample, the relationship between the distribution pattern of heavy metals in the wastewater and leaching properties could also be speculated. In the Pb and Sb samples, the uniformly distributed heavy metals throughout the cement mortar affected active metal leaching. In contrast, Cd is expected to form insoluble compounds within the waste matrix, resulting in leaching resistance in cement wasteform. Nevertheless, these assumptions and the reaction between heavy metals and cement require further investigation to provide a more comprehensive understanding of their behavior within cement-solidified mixed waste.

4. Conclusion

Our study investigated the leaching behavior of heavy metals, including Pb, Cd, and Sb, in cement solidified wasteforms using the mixed waste leaching test standards, TCLP, and ANS 16.1. The cementitious wastes with heavy metals were subjected to additional leaching solutions to simulate the environmental conditions of a silo-type repository. The results showed that Pb and Sb demonstrated increased leachability within silo-type radioactive waste disposal facilities, indicating that the leachability standards for mixed waste in trench-type repositories may not be appropriate for silo-type disposal facilities. Conversely, Cd was not leached in any experimental conditions within the cement wasteform, suggesting the possibility of flexible leaching standards for Cd-containing mixed waste in silo-type disposal facilities. Furthermore, the study revealed that the immobilization effect of cementation on Pb and Sb might be limited, as both metals were observed to dissolve in the cement matrix during the hydration process. In contrast, Cd was partially distributed in the cement mortar, forming agglomerated particles expected to form insoluble compounds. This finding indicates relatively stable solidification and higher leaching resistance of Cd in silo-type repository conditions. Overall, the results highlight the importance of further research to establish appropriate disposal criteria for the environmental conditions of the disposal facilities. Moreover, it is essential to develop adequate treatment techniques for each type of mixed waste to minimize the risk from the leaching of these substances and ensure safe and sustainable waste management practices.

Funding

This work was supported by the Nuclear Safety Research Program through the Korea Foundation of Nuclear Safety (KoFONS), granted financial resources from the Nuclear Safety and Security Commission (NSSC), Republic of Korea (No. 2003016) and KOREA HYDRO & NUCLEAR POWER CO., LTD (No. 2020-Tech-13).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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