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Engineering Measures for Isolation and Sequestration of Heavy Metals in Waste as Safe Final Sink

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Abstract

The long-term safety management of hazardous substances is essential to the development of an environmentally sound resource circulation society. To achieve this, engineering measures to attenuate environmental risks in the isolation and sequestration of hazardous heavy metals are reviewed. From the standpoint of the isolation and sequestration of heavy metals from resource circulation, we assess the challenges in implementing immobilization technologies, constructing updated isolation structures, and controlling environmental conditions. It is also focused intensively on the (bio) chemical transformation behavior of heavy metals and its effect on the migration of the transformed materials in the environment. The contributions of solubilized and gasified metal components to emission into the environment are considered. The obtained results underscore the necessity of multiple barriers to retard and attenuate the migration of hazardous heavy metals. Innovative schemes for the isolation and sequestration of heavy metals will lead to higher levels of safety and environmentally sound resource circulation.

Keywords: safe resource circulation, multi-barrier approach, final sink, heavy metals, immobilization, retarding migration, adsorption

1. Introduction

The world is facing several global and local problems caused by imbalanced resource utilization and inappropriate handling of waste. Climate change is threatening the lives of vulnerable people and regions by increasing extreme weather events. Marine plastic litter is widely recognized as a major risk to maritime activity, fisheries, and wildlife. Waste management practices, which are regarded as one of the major causes of these problems, must be updated to improve the situation.

Achieving environmentally sound resource circulation is a possible solution. The core concept of environmentally sound resource circulation is the harmonization of industrial (human) society with the earth's natural circulation system. It includes measures, such as (a) increased resource circulation to slow down and ultimately close the resource loop by reducing new resource inputs, (b) ethical production of goods

and services that generate hardly any waste to narrow and dematerialize the resource loop, and (c) removal of hazardous substances from the resource circulation loop for the safe loop of resources. Toxic-free resource circulation is essential for the sustainable operation of environmentally sound resource circulation. While it will be achieved, the amount of waste/residue generated can be minimized, and hazards will be removed from the loop of circulation. This minimized residue containing concentrated hazardous substances must be adequately managed to end the negative aspects of these resources.

This chapter describes an engineering approach to isolate and sequester hazardous substances concentrated in waste to remove them from the circulation loop, which will be essential to achieve environmentally sound resource circulation.

2. Concept of sequestration/isolation in environmentally sound resource circulation

The main objectives of the sequestration/isolation of hazardous substances are to reduce and avoid the long-term exposure of humans and the environment to hazardous substances. Environmentally sound resource circulation entails challenges for the actors involved, such as requirements that manufacturers implement cleaner and resource-efficient production. It also means customers should purchase long-life products and recycle their waste. Recycling activity should include only the safe circulation of resources, and hazardous substances must be sequestered from human activity.

The scheme of sequestration of hazardous substances should be consistent with the strategy for a final sink [1]. A final sink is a process in a manner that satisfies the acceptance level of a substance flow as low as the natural environment and the acceptance level of exposure of substances for human health. Hazardous organic chemicals are not expected to be mineralized, and it is hard to secure their long-term immobilization in an isolation site (or a landfill to be upgraded for sequestration). Therefore, the final sink for these substances must occur through physical or chemical destruction. This indicates that the potential of an isolation site to be a final sink is limited to toxic heavy metals or inorganic substances.

Isolation/sequestration should be designed under the multi-barrier approach. The major elements of this approach are (i) containment of toxic substances by stabilization and insolubilization with chemical or physical measures, (ii) avoidance of the release of toxic substances from containment, including exposure to moisture, (iii) retardation of the migration of hazardous substances within a site and the environment, and (iv) early warning of the potential release of a toxic substance by monitoring of containment structures, gases/leachates, and the environment.

3. Containment of toxic heavy metals for safe sequestration

Most countries have legal regulations for the disposal of waste containing hazardous heavy metals. Waste that meets certain criteria can be disposed of in landfills equipped with emission control measures (e.g., leachate treatments and gas collection) [2, 3]. Otherwise, those waste products must be delivered to facilities with containment functions. Pretreatment to detoxify and immobilize waste containing heavy metals is an essential measure to reduce potential emissions reasonably.

Air pollution control (APC) residues, which are generated through thermal treatments of waste, such as incineration, gasification, and pyrolysis, are commonly classified as hazardous materials owing to the high leaching potential of toxic metals. Before APC residues are disposed of, pretreatments are required in many countries to prevent the release of toxic metals into the environment [4]. The available pretreatments can be categorized into three groups—(i) physical or chemical separation, (ii) solidification/stabilization (S/S), and (iii) thermal treatment [5]. Chemical stabilization using organic chelating agents, such as piperazine-based or dithiocarbamate-based agents, is often preferred because such treatments are simple, do not require pretreatments, such as pH control, and remain stable across a wide pH range [6, 7]. On the other hand, these treatments are significantly more expensive than other forms of chemical stabilization [8]. In addition, organic components derived from chelating agents induce long-term leachate problems treatments at landfill sites [9].

3.1 Cement solidification

Cement solidification is a widely used containment technique around the world. The purpose is to avoid the leakage of toxic substances into the environment. The target chemical substances are diverse, such as heavy metals, F, B, and even radioactive substances. Hiraoka and Takeda [10] investigated the effects of cement solidification on the leaching amounts of Hg and Cd in waste sludges in relation to compressive strength. They suggested that the solidification of landfill waste containing heavy metals is safe when the cement amount is over 150 kg/m^3 and the compressive strength is over 0.98 MPa.

Cement solidification was also applied to radioactive cesium-contaminated APC residue generated after the Fukushima Daiichi nuclear disaster in 2011. Radioactive cesium is hardly precipitated in the alkaline condition in a cement mixture and cannot be chemically stabilized, although the solubilities of heavy metals are reduced in alkaline conditions (Figure 1). To reduce the leachability of radioactive Cs, cement solidification should be prepared on a large scale with a value of 1 m^3 so that the specific surface area contacting water is limited. Another improvement is the use of blast furnace cement rather than ordinary Portland cement. Since cement includes Cr as a material component, solidified pieces can potentially leach hexavalent chromium.

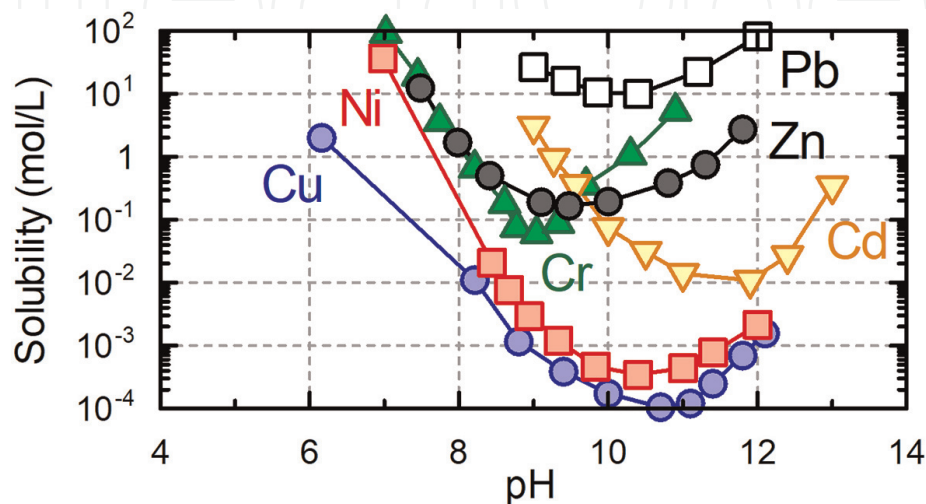


Figure 1.
Relationship between solubility and pH for heavy metal [11].

Blast furnace cement can provide a reductive condition, resulting in the transformation from hexavalent to trivalent chromium leaching. In addition, blast furnace cement is effective for reducing volume expansion and maintaining the long-term containment performance of cement solidifications. APC residue complicates the solidification mechanism of cement because of coexisting reactive chemicals, such as calcium, aluminum, and sulfur. Their chemical components contribute to form an expansive mineral known as ettringite. Volume expansion due to ettringite will generate cracks on the surfaces of solidified pieces, thus increasing the specific surface area. The reduction of both specific surface area and volume expansion is an essential design criterion for controlling the cement solidification of hazardous wastes.

3.2 Solidification by magnesium oxide

Magnesium oxide (MgO) is also an effective binder for solidifying wastes containing heavy metals. There are two methods of producing MgO. One is to bake the natural magnesium carbonate included in dolomite and then crush it. The other is to precipitate Mg ions in seawater as hydroxides and then dehydrate them at high temperatures. This means that magnesium oxide is a safe insolubilizer free from toxic chemicals, which is remarkably different from cement-containing Cr.

When MgO is dissolved in water, magnesium hydroxide is precipitated so that the pH reaches around 10.5 at an equilibrium state. At this pH level, some heavy metals can exhibit the lowest solubility, as shown in **Figure 1**. This has been considered a reason why MgO has a greater ability than cement to immobilize heavy metals. On the other hand, MgO cannot give such a large compressive strength to solidified pieces compared with cement, and MgO is about 8–10 times more expensive than cement. Therefore, it is necessary to optimize the amount and field of usage of MgO. **Figure 2** shows the results of batch leaching tests using an APC residue solidified with blast furnace cement or magnesium oxide. The ratio of ash (A) to binder (B) in weight is parametrically changed. Solidification remarkably has reduced the leachability of Cd, Zn, and F compared with raw APC residue. However, the leachability of Pb cannot be reduced by cement solidification even by increasing the amount of cement. In contrast, magnesium oxide can reduce the leaching amount under specified conditions.

3.3 Stabilization/solidification of mercury-containing waste

According to the framework of the Minamata Convention, the national scheme for the appropriate disposal of Hg-containing waste is required. Due to its environmental effects, Hg-containing waste must be stabilized prior to disposal in HgS form and/or solidified with a polymer or cement to reduce leaching and volatilization. **Figures 3** and **4** show the long-term leaching and volatilization behaviors of processed mercury. Hg-containing waste specimens are first stabilized with sulfide as metacinnabar [13, 14], which has extremely low solubility in water. The specimens are then solidified with one of four binders—sulfur polymer (SP), low-alkaline cement A (CA), low-alkaline cement B (CB), or low-alkaline cement B containing a water-reducing agent (CB+). Here, low-alkaline cement A has hauynite as the main component, and low-alkaline cement B has high-volume fly ash and silica fume. Stabilized Hg-consisting waste solidified with a sulfur polymer exhibits the lowest leaching and volatilization. Low-alkaline-cement-based binders effectively confine Hg but have lower performance than sulfur polymer. pH may significantly affect Hg leachability [15]. Leaching from a piece solidified by low-alkaline-cement binders increases under acidic

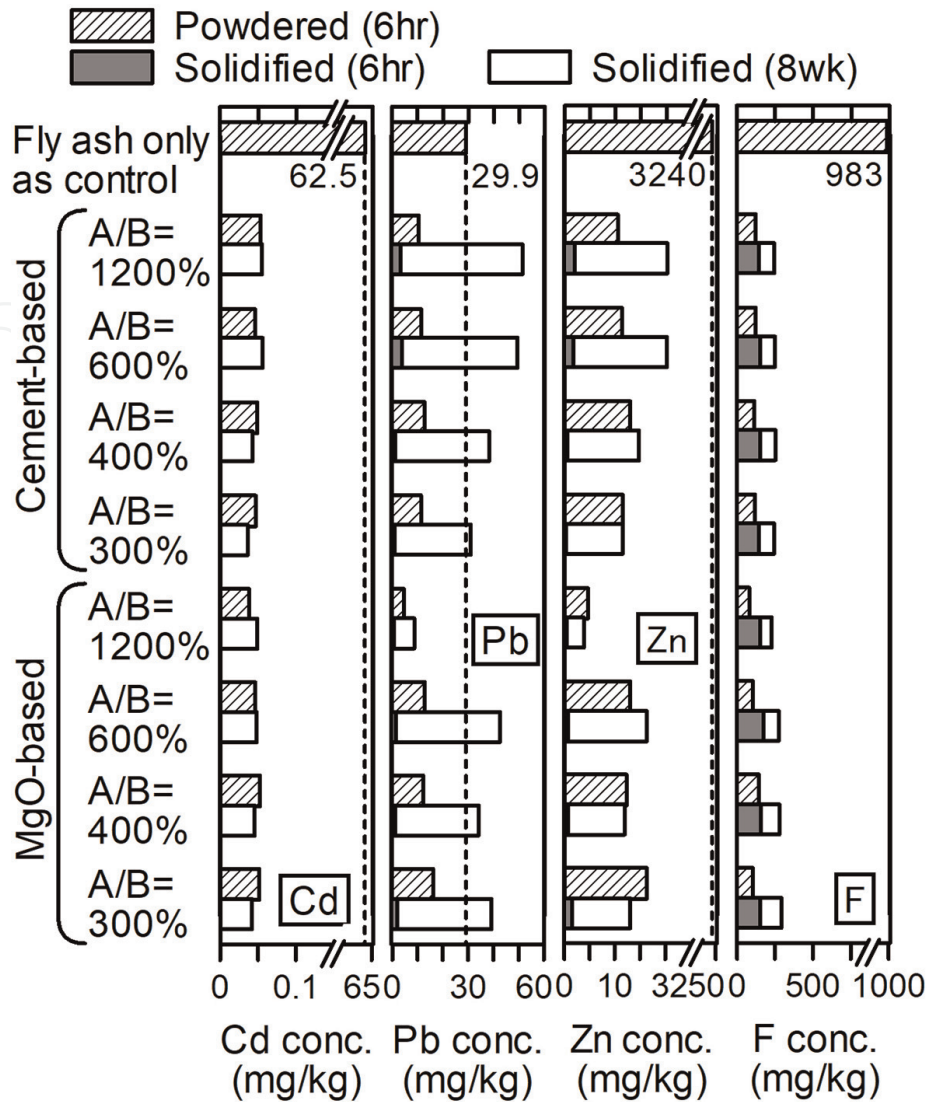


Figure 2. Results of batch leaching tests using an APC residue solidified with blast furnace cement or magnesium oxide [12].

conditions, whereas that solidified with sulfur polymer increases under alkaline conditions. On the other hand, Hg volatilization increases with temperature except for waste solidified with sulfur polymer. Sulfur polymer is effective for decreasing the volatilization rate due to elevated temperature.

3.4 Stabilization by diatomite addition

Among available pretreatments, cement-based S/S is commonly used worldwide [16, 17]. In this process, calcium-silicate-hydrate (C-S-H) gel is formed by the reaction between amorphous silica ($\text{SiO}_2 \cdot n\text{H}_2\text{O}$) and calcium hydroxide ($\text{Ca}(\text{OH})_2$) (pozzolanic reaction) in the cement [18]. Toxic metals can be immobilized by the C-S-H gel via sorption, incorporation, and encapsulation owing to the high microporosity and high surface area [19, 20]. APC residues usually contain high amounts of Ca as a sorbent and reactant for the removal of acidic components in exhaust gas [21]. APC residues often show high pH due to the presence of alkaline Ca compounds [22, 23], and the solubility of amorphous silica increases at alkaline pH [24]. Owing to the high Ca content and alkaline pH provided by APC residues, the addition of amorphous silica to APC residues may induce C-S-H gel formation via pozzolanic reactions for

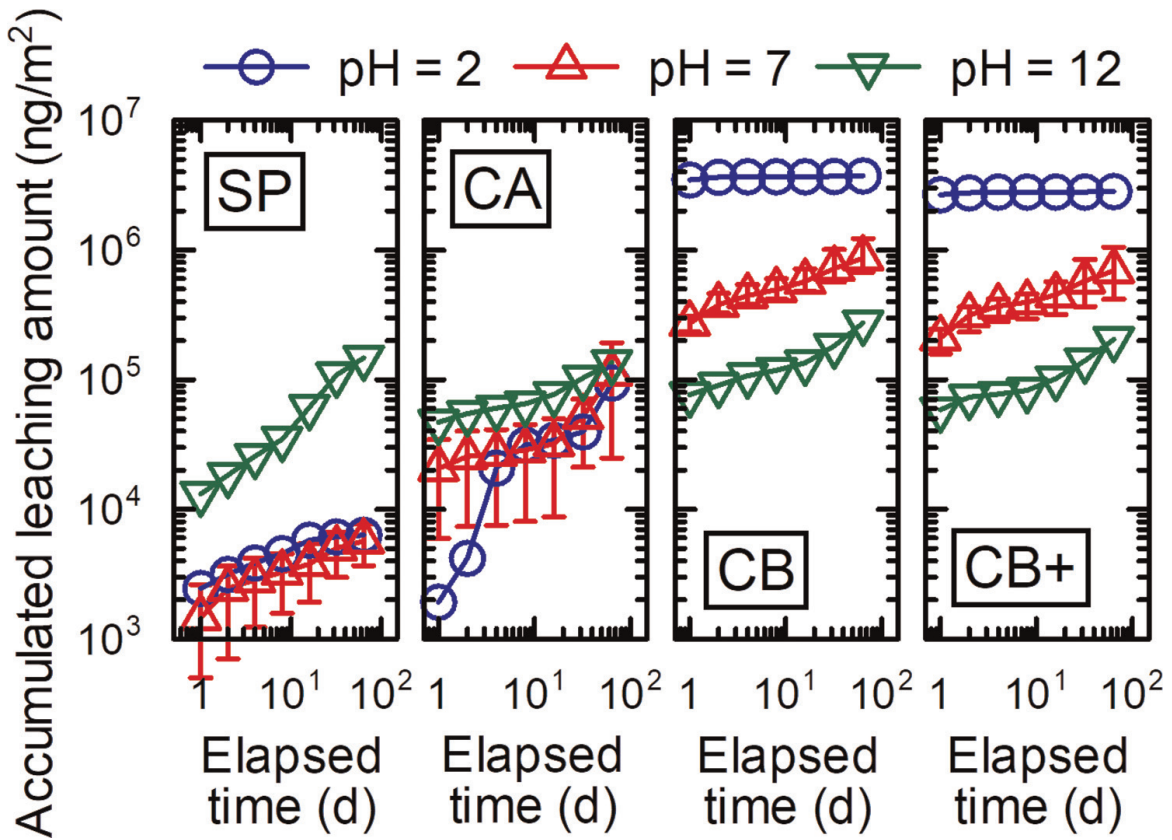


Figure 3. Results of long-term leaching tests: Effects of pH on Hg leaching.

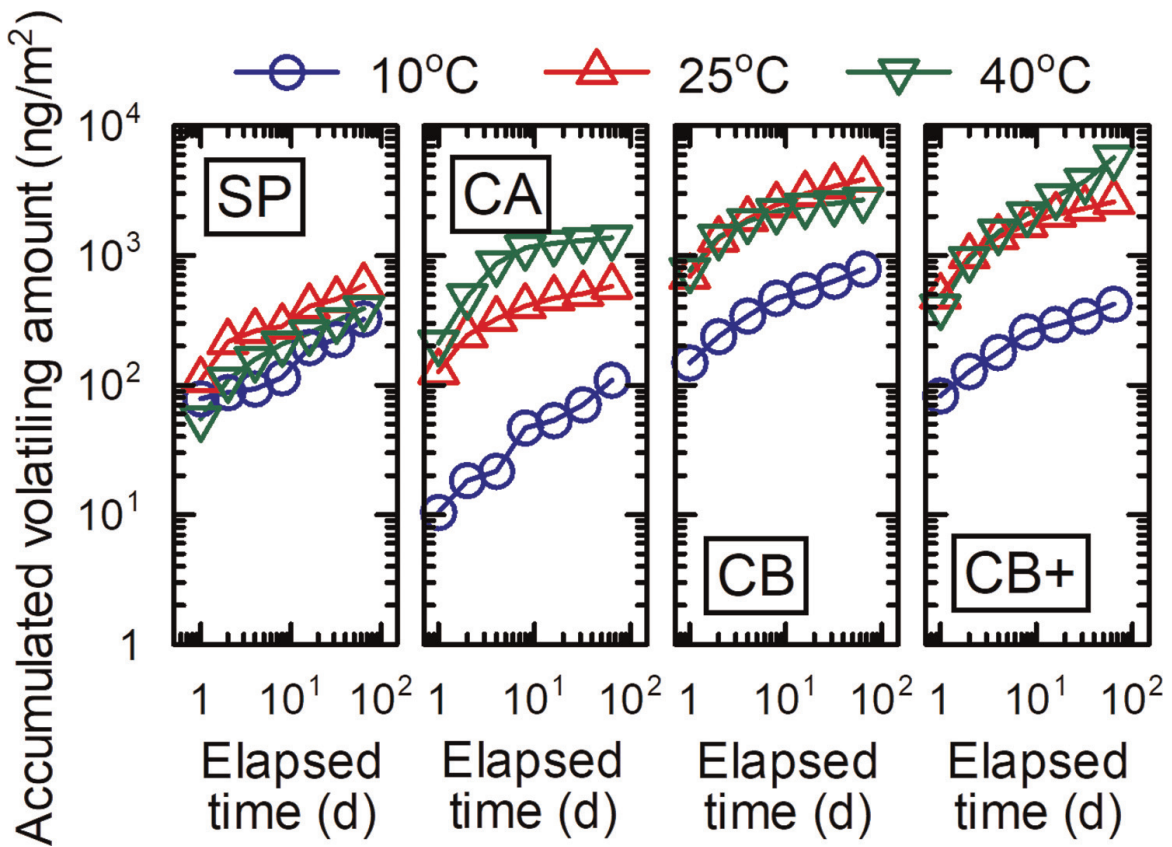


Figure 4. Results of long-term volatilization tests: Effects of temperature on the accumulated amount of volatilized Hg.

metal immobilization. Thus, both the treatment cost and the use of chemical agents might be reduced by using inexpensive silicon materials instead of cement. We considered diatomite as a natural pozzolanic additive [25] for lead immobilization in APC residues owing to its high amorphous silica content [26, 27], relative abundance [28], and low cost compared to Portland cement [29].

Assessment of the impact of diatomite addition on Pb immobilization in APC residues (**Figure 5**) indicates that Pb leaching from weathered APC residues decreased as time and temperature increased. This is attributed to the increase in the hydration reaction of cementitious materials as the temperature increases [30, 31]. At each weathering temperature, Pb leaching from stabilized APC residues decreased as diatomite doses increased. The leaching amount of Pb from 14-day stabilized APC residues with 0%, 5%, or 10% diatomite addition was reduced by 18–67%, 67–90%, or 80–99%, respectively. Consequently, the leaching amount of Pb dropped below 0.3 mg/L (Japanese criterion for landfill disposal) after 14 days of curing with the addition of 10% diatomite at 70°C.

Figure 6 shows the X-ray diffraction (XRD) patterns of raw APC residues and 14-day cured APC residues following the addition of 10% diatomite at 70°C. The peak

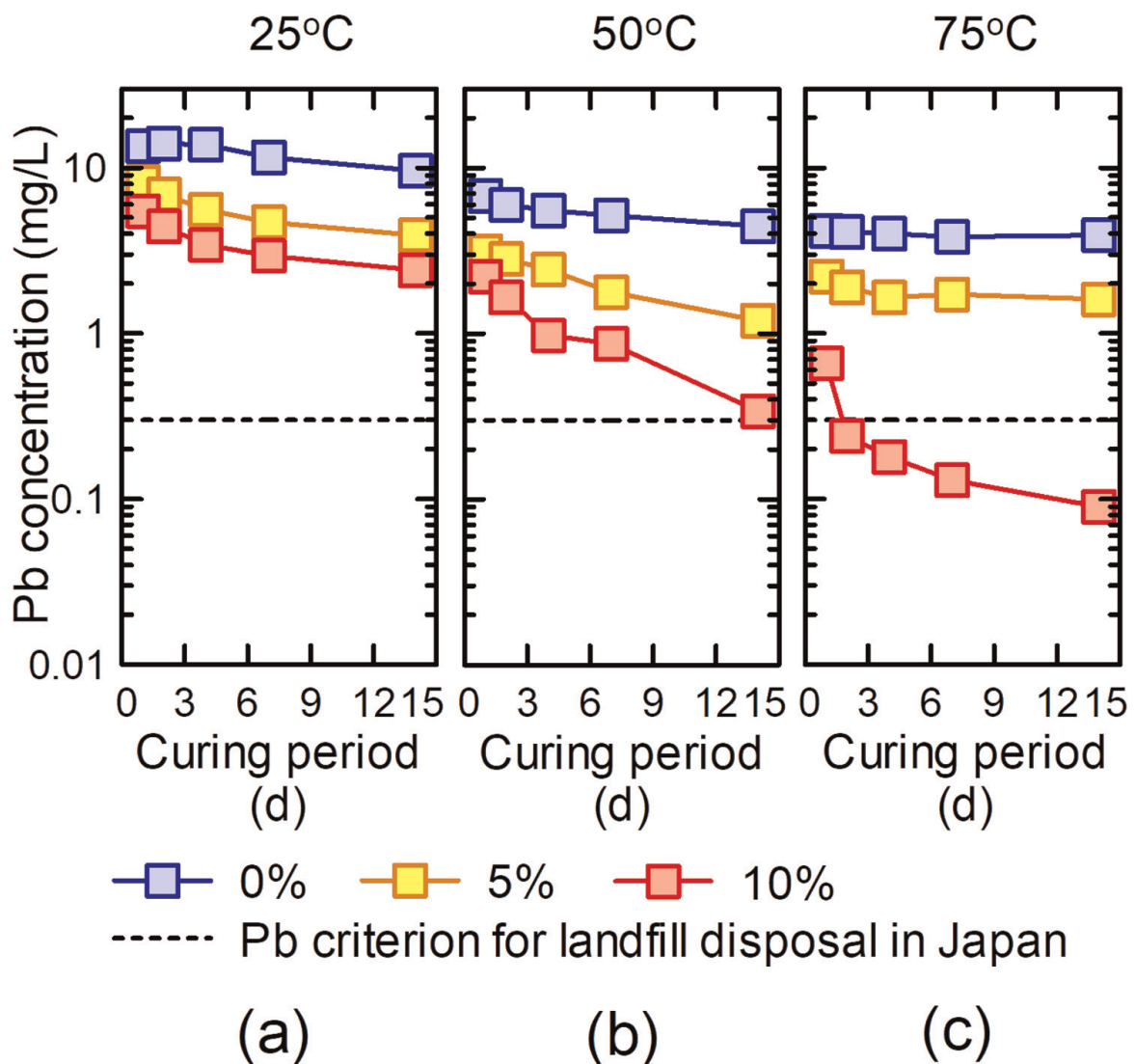


Figure 5. Leaching concentrations of Pb from cured APC residues. APC residue under the temperature of (a) 25°C, (b) 50°C, (c) 75°C.

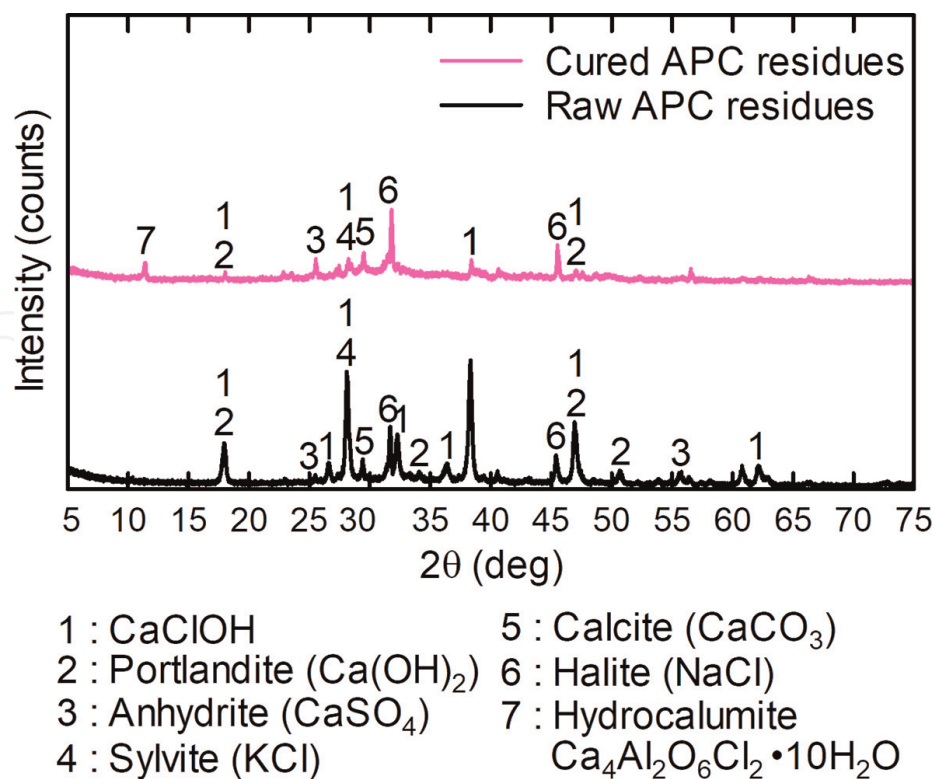


Figure 6. XRD patterns of raw and cured APC residues by 10% of diatomite at 70°C for 14 days.

intensities of CaClOH and Ca(OH)₂ significantly decreased by weathering with diatomite, indicating that they reacted to diatomite and were consumed by the **Figure 6** XRD patterns of raw and cured APC residues by 10% of diatomite at 70°C for 14 days pozzolanic reaction. New peaks of C–S–H gel in the stabilized APC residues were not confirmed, as the residues were below the detectable level of XRD analysis or crystallization was incomplete [32]. Even though C–S–H gel formation was not detected in the XRD analysis, the amount was sufficient to immobilize 99% Pb in the APC residues.

Diatomite, consisting mainly of amorphous silica, was used as a pozzolanic additive for Pb immobilization in APC residues instead of cement. The results showed that the leaching amount of Pb from the stabilized APC residues was reduced by C–S–H gel formation via the pozzolanic reaction among Ca(OH)₂, CaClOH, and diatomite. Consequently, the leaching amount of Pb dropped below 0.3 mg/L. This study showed the feasibility of using diatomite to immobilize Pb in APC residues. Although 10% diatomite was added to the APC residues, the volume increase is supposed to be lower than that in cement-based S/S [33]. From the viewpoint of landfill management, this treatment would reduce the use of chelating agents while suppressing the increase in volume. If wastes containing amorphous silica can be used to immobilize metals in APC residues, this method has the potential to be a low cost and environmentally friendly solution.

4. Weathering and attenuation of waste in landfills: impact of mineral compositions on metal leachability

Waste containing toxic metals, which are commonly immobilized before disposal, will be stabilized by a chemical or microbial weathering process in landfills. For example, precipitation of insoluble metal sulfide in the presence of sulfides produced

by sulfate-reducing bacteria is one of the major mechanisms by which toxic metals are immobilized in landfills [34]. In the initial stage of the weathering process, calcite (CaCO_3) generated by carbonation via atmospheric CO_2 immobilizes toxic metals by absorption on the surface and/or incorporation into the crystal structure [35, 36]. Thus, these processes have non-negligible impacts on metal mobilities at waste landfills. In this context, the impacts of mineral compositions of waste in a landfill on the leaching behaviors of heavy metals were analyzed. The obtained waste was composed of the following—cover soils (0–0.15 m), waste layers (0.15–47.6 m), embankment (47.6–50.9 m), and gravel layers (50.9 m–56 m). Mineral compositions identified by XRD analysis (**Table 1**) showed that the waste in this landfill mainly consisted of calcite (CaCO_3), gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), and quartz (SiO_2). The contents of heavy metals (Cu, Mn, Ni, and Zn) and their leachability (**Figure 7**) revealed that the waste at 31.3 m depth had high contents and high leaching concentrations of Cu, Mn, Ni, and Zn. The core sample contained weddellite ($\text{CaC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$) and whewellite ($\text{CaC}_2\text{O}_4 \cdot \text{H}_2\text{O}$). Waste incineration residues are known to contain small amounts of organic acids, such as oxalic acid ($\text{C}_2\text{H}_2\text{O}_4$) [37]. Calcium oxalate (weddellite and/or whewellite) can be formed in waste incineration residues by the coprecipitation of Ca^{2+} ion and oxalic acid [36]. It is also considered that calcium oxalate in sludge produced by the neutralization of waste acid using calcium hydroxide has been landfilled. Another possible source of calcium oxalate is waste from the ceramics industry. The waste also contains iron oxide (see **Table 1**) that, like calcium oxalate, is used for ceramic glazes. Thus, oxalic acid and calcium oxalate could be in the waste. If oxalic acid has a high capability of extracting heavy metals, it might enhance the leachability of heavy metals in waste landfills. Compared to mineral acids, the contents of oxalic acid (organic acid) do not seem high, owing to its precipitation as a metal–oxalate complex [22]. On the other hand, the mobility of heavy metals seems to be significant in this landfill according to the solubility of the metal–oxalate complex (**Table 2**) [38]. Moreover, the stability constant of this complex is higher than that of calcium oxalate (**Table 2**), suggesting that the mobility of heavy metals might be enhanced by the

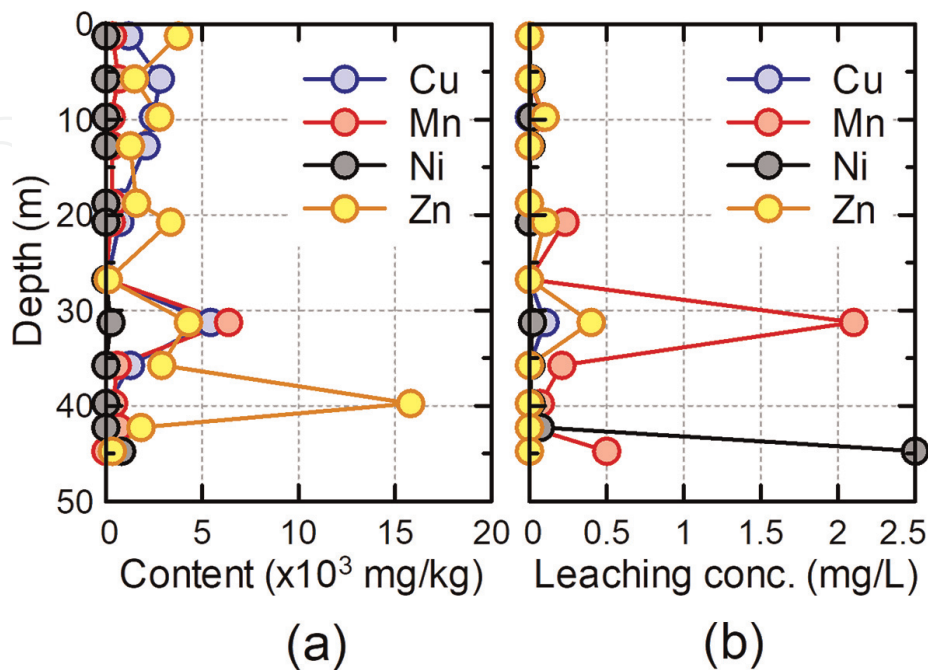


Figure 7. Contents and leaching concentrations of heavy metals in waste. (a) contents, (b) leaching concentrations.

Depth (m)	Albite	Calcite	Chlorite	Gibbsite	Gypsum	Iron oxide	Muscovite	Quartz	Weddellite	Whewellite
1.25		+			+			+		
5.75		+			+			+		
9.75		+			+			+		
12.75		+			+			+		
18.75		+						+		
20.75		+			+			+		
26.75		+						+		
31.25		+			+	+		+	+	+
35.75		+						+	+	+
39.75	+	+	+				+	+		
42.25		+			+			+		
44.75				+	+			+		
					+: Presence					

Table 1.
Mineral compositions of a waste obtained from the landfill.

Metal	Ca ²⁺	Cu ²⁺	Zn ²⁺	Pb ²⁺
Solubility in water (mg/100 mL)	0.67	2.53	0.79	0.16
Stability constant	3.00	6.25	4.87	4.91

Table 2.
Solubility and stability constant of metal oxalate in water.

formation of a metal–oxalate complex via a substitution reaction between Ca²⁺ ion in calcium oxalate and divalent metals.

Our assessment revealed the high leachability of Cu, Mn, Ni, and Zn from waste containing calcium oxalate. Calcium oxalate could be in waste from possible sources, such as incineration residues, sludge produced by waste acid treatment, and ceramics. This suggests that the existence of specific minerals, such as calcium oxalate (weddelite and/or whewellite), might enhance the leachability or mobility of heavy metals in landfills to some extent.

5. Soil adsorption barrier for retarding transport of heavy metals

5.1 Distribution coefficients

The soil adsorption performance of heavy metals, which directly affect migration, has been of great interest among engineers. Many reports about adsorption parameters for various soil types have been published. In particular, the Japan Atomic Energy Agency (JAEA) has established a database summarizing the results of soil adsorption tests using radioactive isotopes [39]. This section overviews soil adsorption parameters (distribution coefficients) of radioactive isotopes, such as Hg, Cd, Pb, Se, and Cs, as references. Cr and As do not have radioactive isotopes and JAEA does not support their distribution coefficients. The distribution coefficients are collected with previous references targeted to soils in major countries (**Figures 8–14**) [40–55].

The distribution coefficients of any heavy metals take a wide range of values. Here, the geometric mean value in each soil type against heavy metals is presented as a representative value in **Figures 8–14**. The distribution coefficient indicates a soil

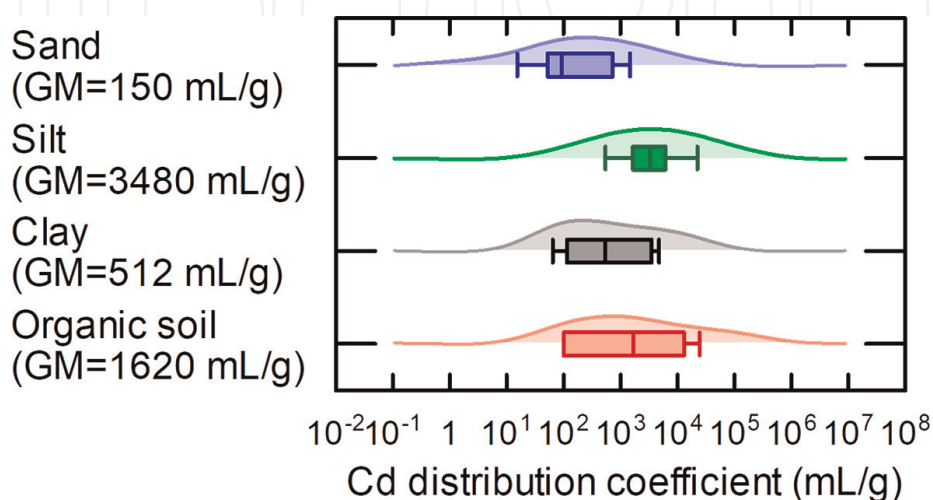


Figure 8.
Distribution coefficients of cadmium arranged from JAEA database.

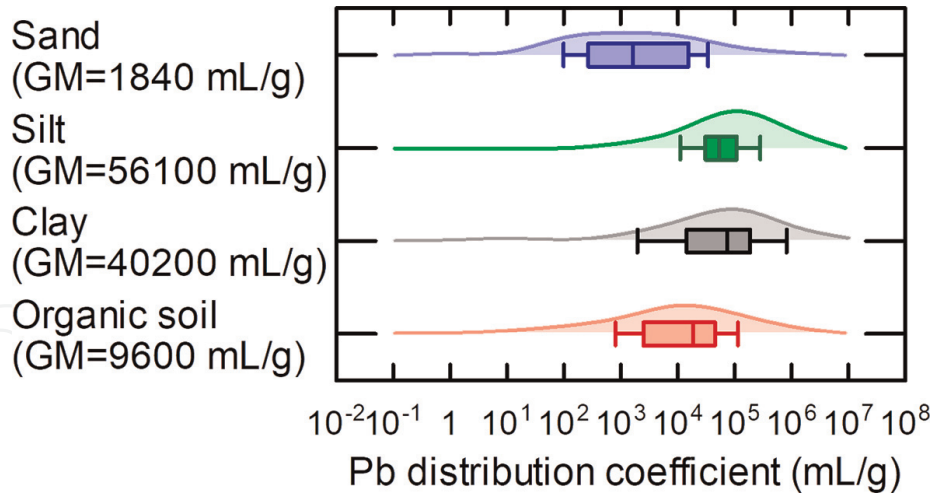


Figure 9.
Distribution coefficients of lead arranged from JAEA database.

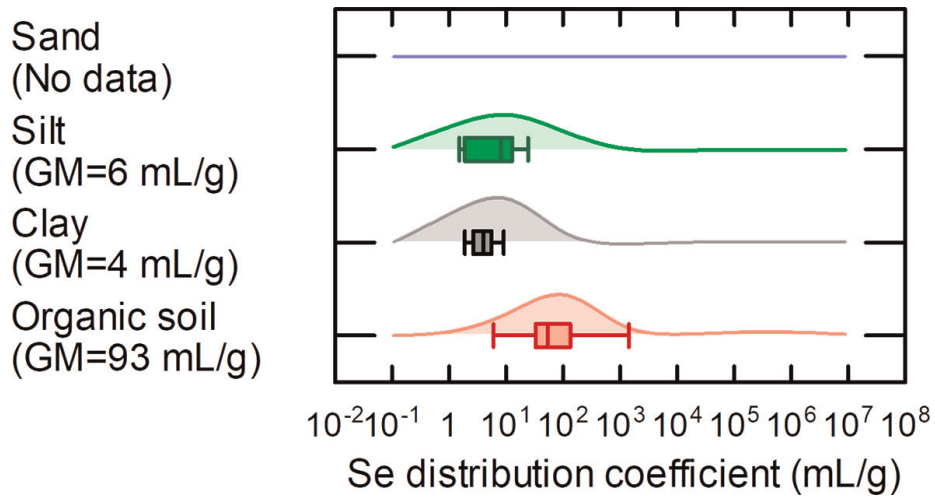


Figure 10.
Distribution coefficients of selenium arranged from JAEA database.

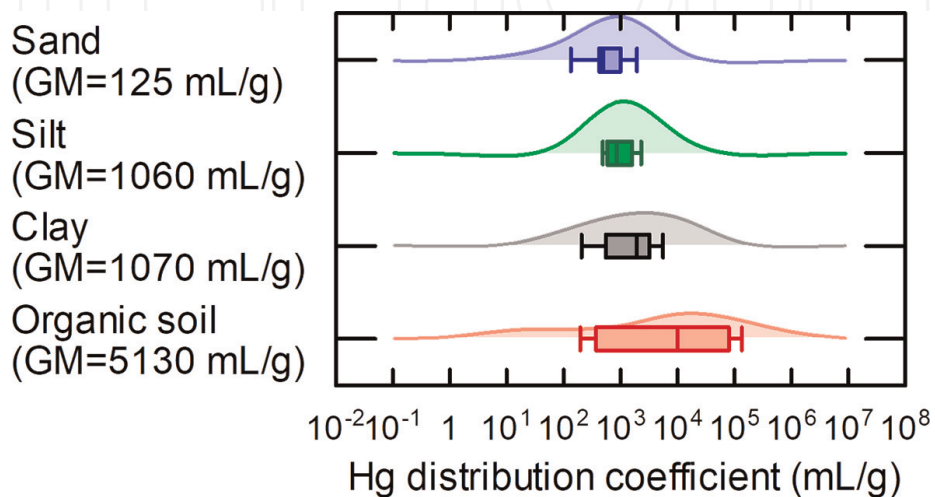


Figure 11.
Distribution coefficients of mercury arranged from JAEA database.

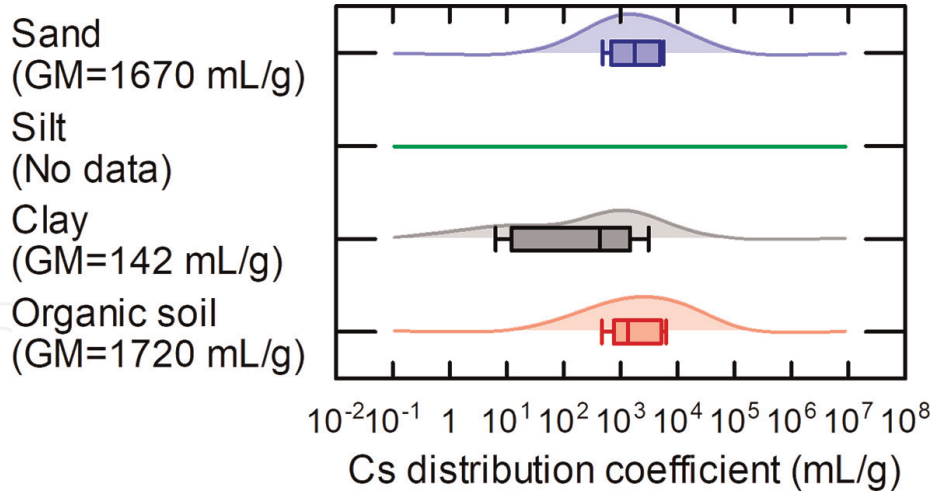


Figure 12.
 Distribution coefficients of cesium arranged from JAEA database.

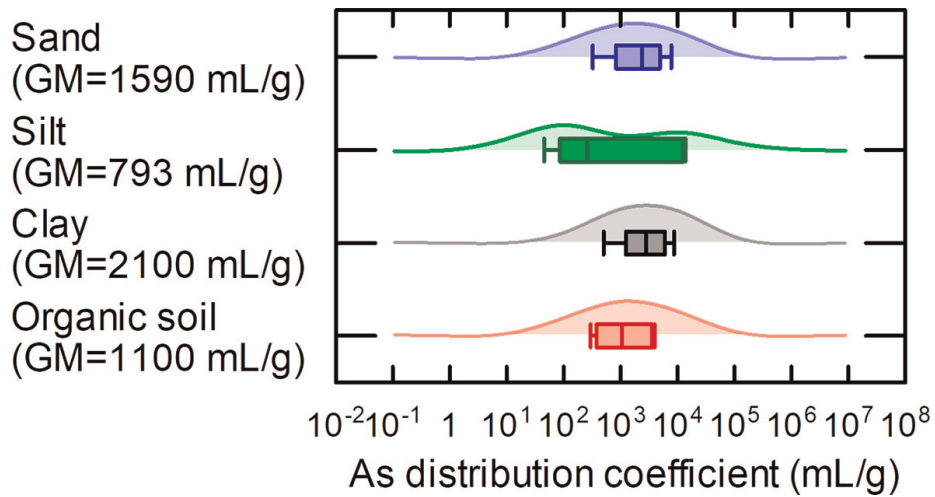


Figure 13.
 Distribution coefficients of trivalent or quinquevalent arsenic.

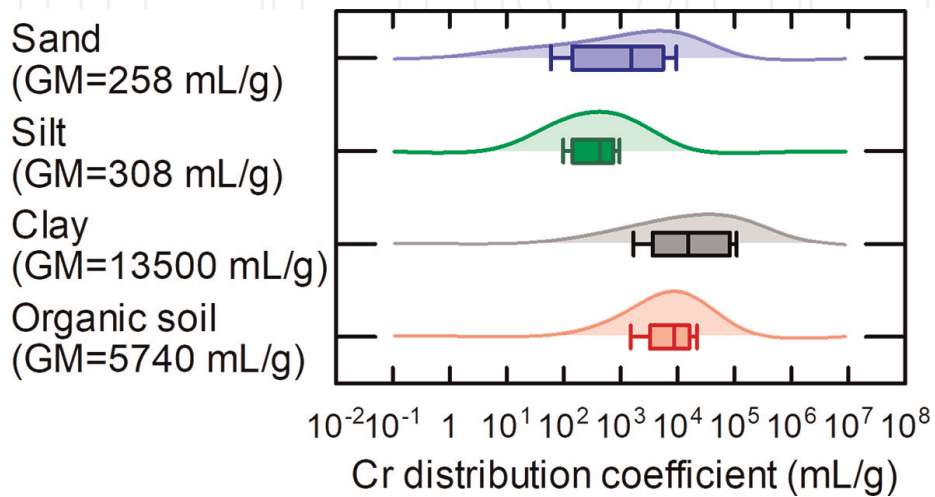


Figure 14.
 Distribution coefficients of trivalent or hexavalent chromium.

type's ability to adsorb a heavy metal. The larger the distribution coefficient, the greater the retardation of the chemical substance transport, resulting in superior barrier performance against the transport. The distribution coefficients against Cd, Pb, Hg, and Cs are relatively high, whereas those against Se, As, and Cr are lower. This is because the chemical forms of Se, As, and Cr in water are anions that are hard to adsorb to the soil surface with a negative charge and whose transport is hard to retard. Therefore, the environmental impacts of the transport of Se, As, and Cr with such small distribution coefficients should be carefully evaluated.

Se, As, and Cr in water can have anionic forms with different ionic valences, depending on environmental conditions, such as pH and oxidation–reduction potential. Numerous review studies, including this chapter, describe in broad strokes the distribution coefficients of heavy metals, but they hardly investigate the differences in distribution coefficients of heavy metals with different ionic valences. However, some scientific papers investigate the effects of ionic valence on distribution coefficients. For example, hexavalent chromium has smaller distribution coefficients than trivalent chromium [54], but both have values in the range of 100 mL/g or less at maximum [55]. Kumpiene et al. [56] review the stabilization mechanisms of As, Cr, Cu, Pb, and Zn. Especially, they discuss the stabilization of As, which is dependent on seven factors—iron compounds, aluminum oxides, manganese oxides, organic matter, alkaline materials, clay minerals, and sulfides. The fact that As can be adsorbed on Fe has been considered a reason why As and Pb included in slags do not leach into water [57–59].

Soil pollution is a global environmental problem. As and Cr are relatively common as causative pollutants, so, their findings are collected and shared among not only researchers but also practitioners. In contrast, Se is a relatively minor substance in soil pollution and waste management, and thus far there are few studies on Se. Further studies on Se, As, and Cr are needed to accurately manage human risk because their distribution coefficients are small and environmental pollution by them is easily spread.

5.2 Soil adsorption barrier for gaseous Hg

Soil adsorption is also effective for retarding gaseous substances and preventing their diffusion. In general, there are three methods for evaluating the distribution coefficients of gaseous substances—(i) dynamic adsorption column, (ii) gravimetry, and (iii) constant volume [60]. The constant volume method has commonly been used to obtain adsorption isotherms. However, gases often adsorb on the container surface or leak from the plug, so obtained isotherm data need to be compensated using the losses of the gases in a blank test. In the previous studies that evaluate the adsorption abilities of adsorbents against some volatile organic compounds (VOCs) using Tedlar bags [61], the compensated isotherms at the equilibrium state can be exactly calculated because the losses of the VOCs in the Tedlar bags are mostly due to adsorption on the surface.

Gaseous Hg, however, would not only adsorb on the container surface but also leak from the plug, so, the equilibrium state cannot be reached. This characteristic of gaseous Hg makes the evaluation of its distribution coefficients difficult. Therefore, a testing method to evaluate adsorption abilities under a nonequilibrium state caused by the leakage should be established.

Ishimori et al. [62] suggested the constant volume method for evaluating the adsorption characteristics of soils and adsorbents against gaseous Hg under a nonequilibrium state. They formulated the phenomenon of nonequilibrium soil adsorption with leakage using the Langmuir sorption model and the diffusive leakage

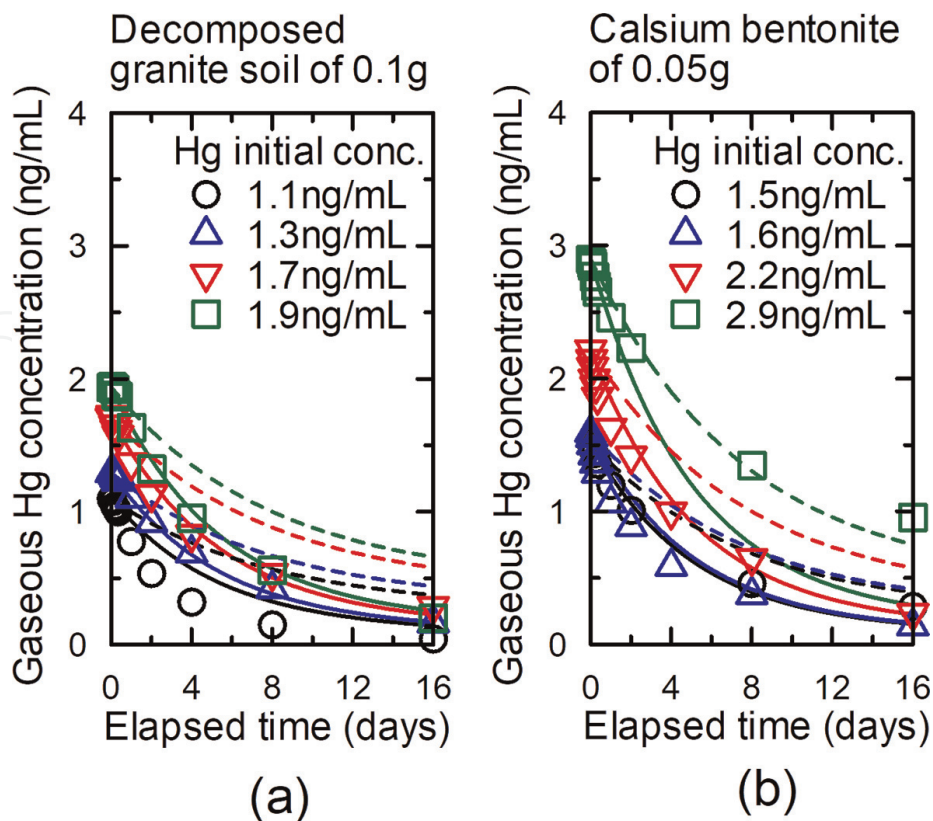


Figure 15. Kinetics of gaseous Hg during adsorption tests for decomposed granite soil (a) and calcium bentonite (b). Plots: Experimental results, solid lines: Fitting results to a governing equation, dashed lines: Estimation results with neither Hg adsorption on container surface nor leakage from sealing plug.

model, resulting in an estimation of distribution coefficients of gaseous mercury by fitting experimental data to the governing equation (see **Figure 15**). Finally, the adsorption isotherms for sand, granite soil, calcium bentonite, and mordenite are estimated as shown in **Figure 16**. Then their distribution coefficients values for gaseous mercury are obtained as 56.3, 2070, 7140, and 3490 mL/g, respectively. It is noted that their values are obtained from the initial slopes of their adsorption isotherms when the equilibrium concentrations are zero.

Hg-coning wastes will be disposed of in landfill sites in the near future due to the signing of the Minamata Convention on Mercury. Soil adsorption barriers are an effective containment method to retard the transport and minimize the emission of mercury. The distribution coefficient is the most important parameter for providing the required containment barrier performance in landfills. The soil adsorption characteristics against both the aqueous and gaseous forms of mercury are insufficiently investigated thus far. It is well known that distribution coefficients depend not only on the type of soil and the adsorbents but also on environmental conditions, such as pH, oxidation–reduction potential, temperature, and coexisting aqueous or gaseous substances.

6. Long-term environmental safety evaluations using multiphysics numerical simulations

Transport of aqueous and gaseous Hg in controlled landfills.

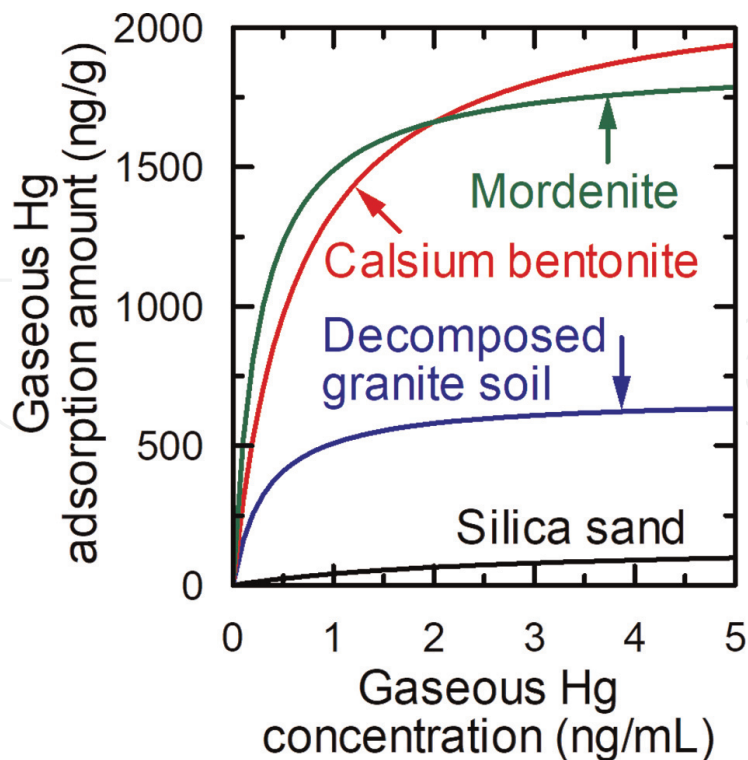


Figure 16. Adsorption isotherms of four samples against gaseous Hg [59].

In environmental engineering, numerical simulation is a method of predicting the fate and transport of chemicals. It is especially effective for evaluating long-term environmental safety, which cannot be predicted in experiments under limited conditions, such as testing duration, the scale of the domain targeted, and its heterogeneity. Numerical simulation can be conducted by solving governing equations that mathematically express the phenomenon targeted by prediction. Governing equations are always formulated to satisfy the law of mass conservation for targeted substances. Optionally, governing equations are also formulated to satisfy the law of momentum or energy conservation. Here, examples of numerical simulations for the transport of aqueous and gaseous mercury in landfills after disposal of mercury-containing waste are introduced, and the effectiveness of those simulations for long-term safety evaluation are discussed.

Ishimori et al. [63] investigated the environmental safety of landfill sites in which Hg-consisting waste is disposed of. Serial batch tests are conducted to evaluate the long-term leaching and volatilization of Hg stabilized in its sulfide form and solidified using either a sulfur polymer or low-alkaline cement. Using measured Hg leaching and volatilization rates, numerical simulations are conducted to investigate the long-term behavior of Hg after its disposal in landfill sites.

Figure 17(a) shows the analysis domain and conditions. A landfill site disposing of Hg-containing waste is modeled in the cross-sectional domain, and the waste is treated through stabilization and solidification techniques as shown in **Figures 3** and **4**. After treatment, the waste takes the form of a 1 m^3 cube. The entire array of solidified cubes is covered with a soil sorption layer, designed to retard the transport of emitted Hg. A drainage pipe is placed at the bottom of the analysis domain to accumulate the leachate, and a final cover or cutoff layer to reduce rainfall permeation is placed on top. **Figure 17(b)** and **(c)** show the initial and boundary conditions of the analysis—the boundary condition at the top of the domain indicates the rainfall

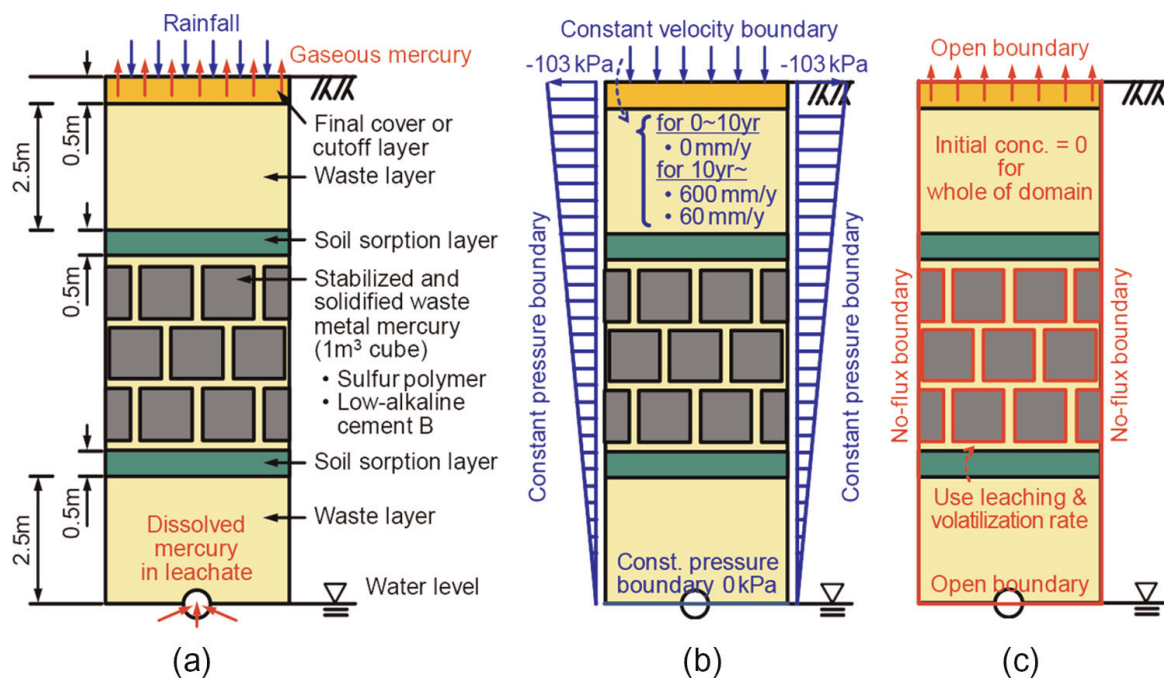


Figure 17. Analysis domain and conditions: (a) two-dimensional cross section of the landfill, (b) initial and boundary conditions for seepage analysis, (c) initial and boundary conditions for advection–dispersion analysis [60].

intensity. For the first 10 years of the analysis, the rainfall intensity for the top boundary condition is considered to be 0 mm/y, as landfilling of the solidified piece would be carried out under a roof; at the end of the landfilling process, the roof would be removed, so in subsequent years, rainfall at the top boundary condition is considered to permeate the final cover with an intensity of either 600 mm/y or 60 mm/y. These different rainfall intensity values are used to evaluate the effects of using a cutoff layer covering the waste site, which would decrease the overall ingress of water to the landfill site. The measured leaching and volatilization rates of mercury are applied to the surface boundaries of the stabilized, solidified Hg-consisting waste.

This numerical simulation investigates the effects of the soil sorption and cutoff layers on the concentrations of dissolved Hg in the leachate at the bottom of the drainage pile and of gaseous Hg emitted from the final cover—the analytical conditions for these models are listed in **Table 3**. These parameters are given to the governing equations. In this study, they are formulated based on the law of mass conservation regarding water, air, dissolved Hg, and gaseous Hg, where the governing equations for water and air are called two-phase flow models, and the governing equations for dissolved and gaseous Hg are called advection–dispersion equations in general. Their equations have been widely used in numerical simulations for fluid flow and chemical substance transport in porous media. A notable point is that the phase transfer rate between dissolved and gaseous Hg is modeled using the Henry constant of Hg. The equation systems mentioned above can be numerically solved using many commercial simulation software programs or open-source codes. The following results are obtained from numerical solutions by COMSOL Multiphysics ver 5.0 (COMSOL, Inc).

A drastic difference between solidification by sulfur polymer and that by low-alkaline cement appears in the total Hg emissions from the landfill (**Figure 18**). The total Hg emission depends significantly on the presence of a soil sorption layer and

Parameters	Unit	Waste layer	Soil sorption layer
Porosity	1	0.3	0.3
Intrinsic permeability	m ²	1 × 10 ⁻¹²	1 × 10 ⁻¹²
Dry bulk density	kg/m ³	1400	1400
VG parameter, α	1/m	2	2
VG parameter, n	1	1	1
Longitudinal dispersivity	m	3	3
Transversal dispersivity	m	1	1
Distribution coefficient	mL/g	0	100
Henry constant	1	0.43	0.43

Table 3.
Analytical conditions for predicting mercury behavior in landfill.

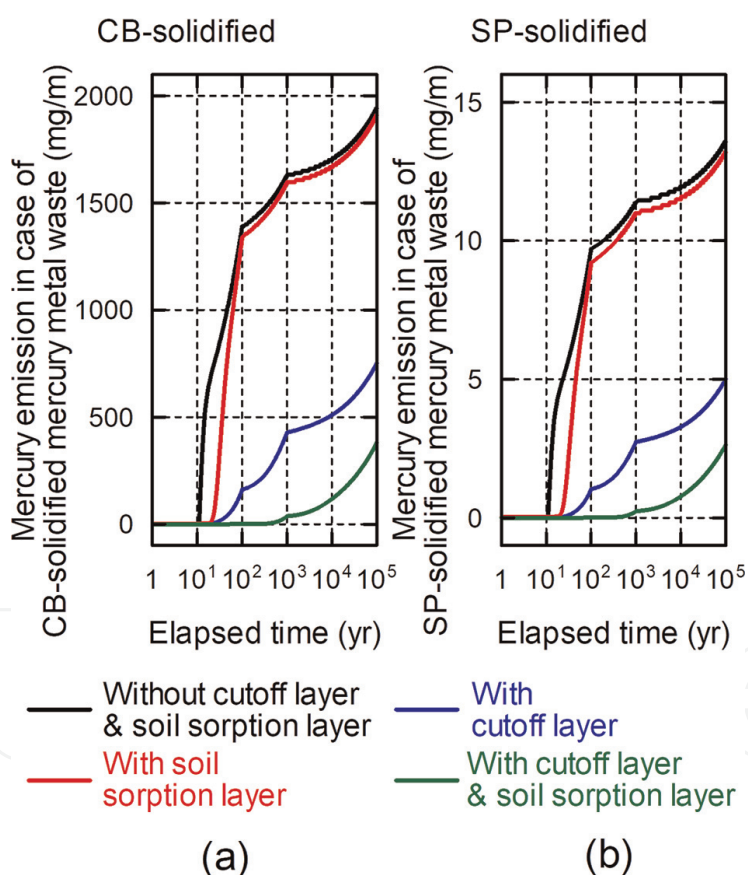


Figure 18.
Total amount of mercury emitted from a landfill with Hg-consisting waste solidified by (a) sulfur polymer or (b) low-alkaline cement.

cutoff layer as well as on types of binders to solidify the Hg-consisting waste. The most effective measure to reduce Hg emission is considered to be sandwiching sulfur polymer-solidified pieces between sorption layers and covering the landfill surface with a cutoff wall. Numerical simulations will help us design the required geometry and material quality of the soil sorption layer, the cutoff wall, and the stabilized solidified Hg-consisting waste.

Hazardous waste containment performance depends on the aging of RC materials or their failure due to chemical attack.

Another practice of numerical simulation is the multiphysics of seismic analysis and reactive chemical transport analysis to evaluate the long-term environmental safety of isolation-type landfills that are designed for hazardous waste and that have a reinforced concrete structure [64]. Hazardous wastes possess a hazard to human health and the environment when improperly managed. They have extremely high leaching concentrations, so they cannot be disposed of directly into regular landfill sites. In general, such hazardous wastes are strictly controlled in waste containment facilities whose function is to prevent penetration and thus to avoid the wastes from leaching due to rainfall [2, 3]. As case studies, this containment facility so-called isolation-type landfills have been built using waterproof reinforced concrete with a thickness > 350 mm and compressive strength >25 MPa, based on the regulations in Japan. Ishimori et al. [65] used numerical simulations to show the importance of a multi-barrier system consisting of stabilization/solidification techniques and artificial/natural soil sorption layers to minimize the negative impacts of a hazardous waste landfill. These numerical simulations consist of seismic analysis to evaluate the stability of the structure in the event of huge earthquakes and reactive chemical transport analysis to predict the long-term leaching concentration profiles from landfills damaged due to deterioration over time or sudden huge earthquakes.

Figure 19 overviews those numerical studies. The analysis domain consists of a waste containment facility and its surrounding grounds. The environmental safety of hazardous waste in the facility, which is built with reinforced concrete, is evaluated by predicting the concentrations of heavy metals at monitoring well located in the lower reaches of the groundwater. Four numerical studies are performed. In Case 1, no earthquake occurs for 100 years. In Cases 2 and 3, a small and a large earthquake, respectively, occur after 5 years. Each earthquake is equivalent to a magnitude of a Level 1 (L1)

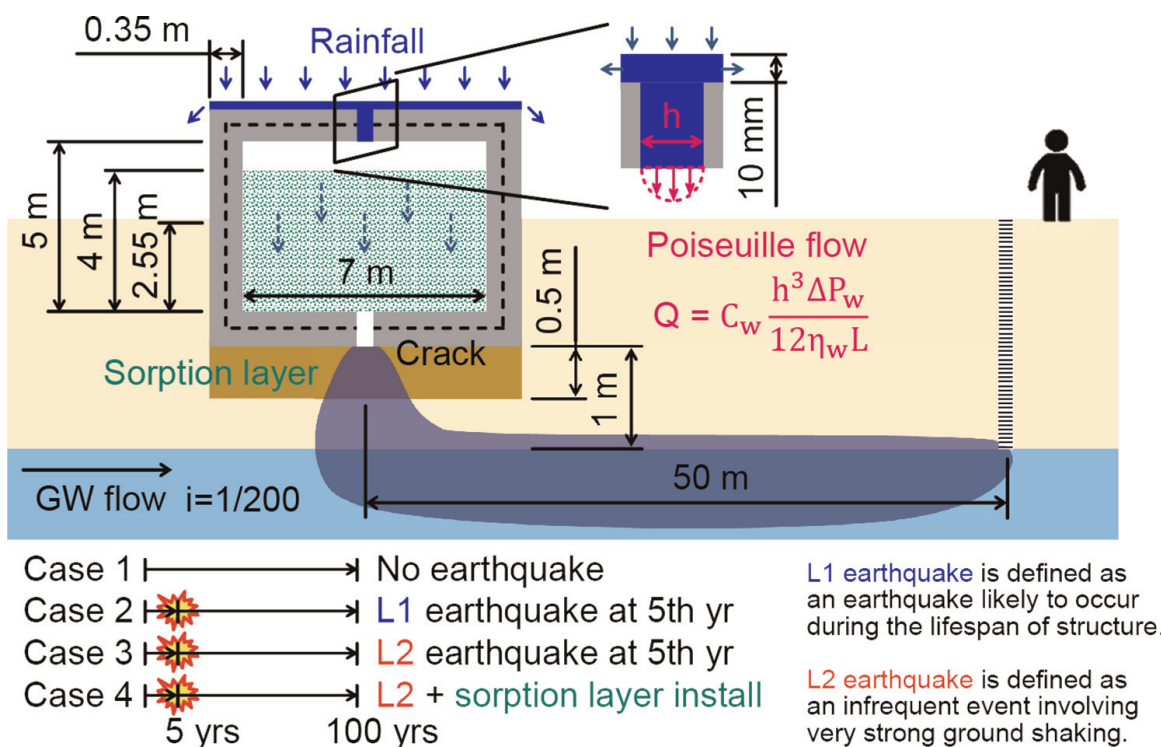


Figure 19.
 Analysis domain and conditions for case studies.

or Level 2 (L2) earthquake as defined in Japan, respectively. Case 4 additionally has a 50-cm sorption layer underlying the waste containment facility. Hazardous waste is assumed to be APC residue, and cadmium is targeted as a contaminant.

Figures 20–22 overview the whole analytical procedure. Table 4 shows the main analytical conditions used. The governing equations are formulated from the law of

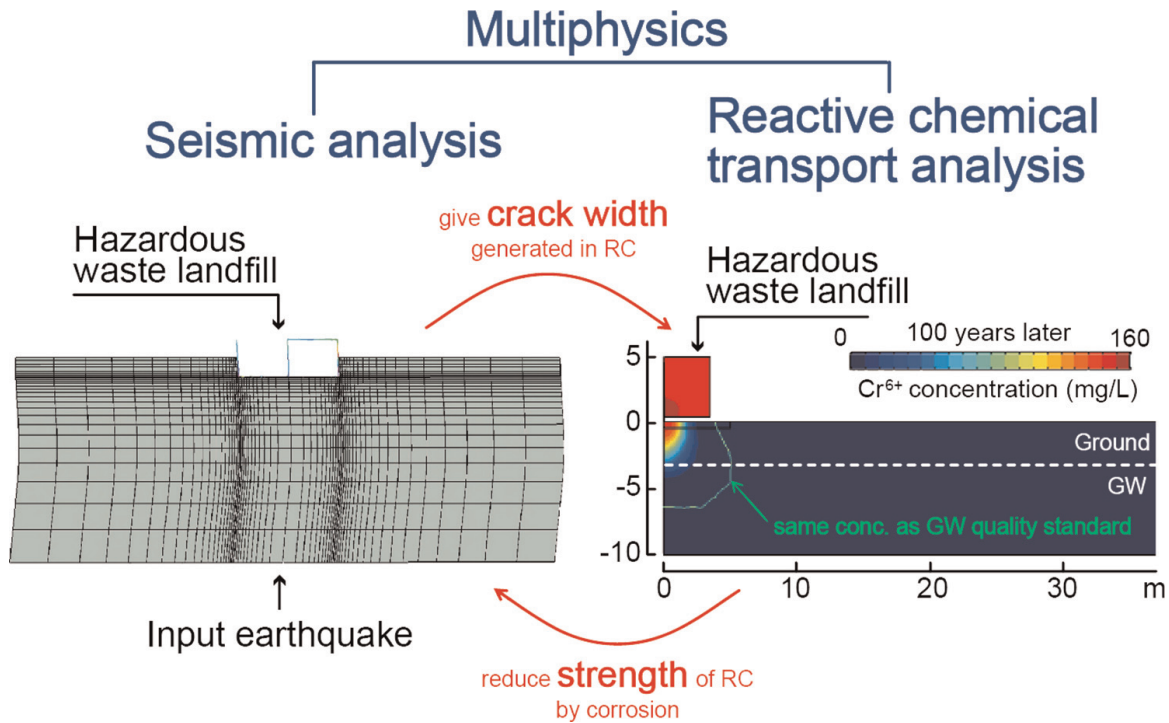


Figure 20. Multiphysics.

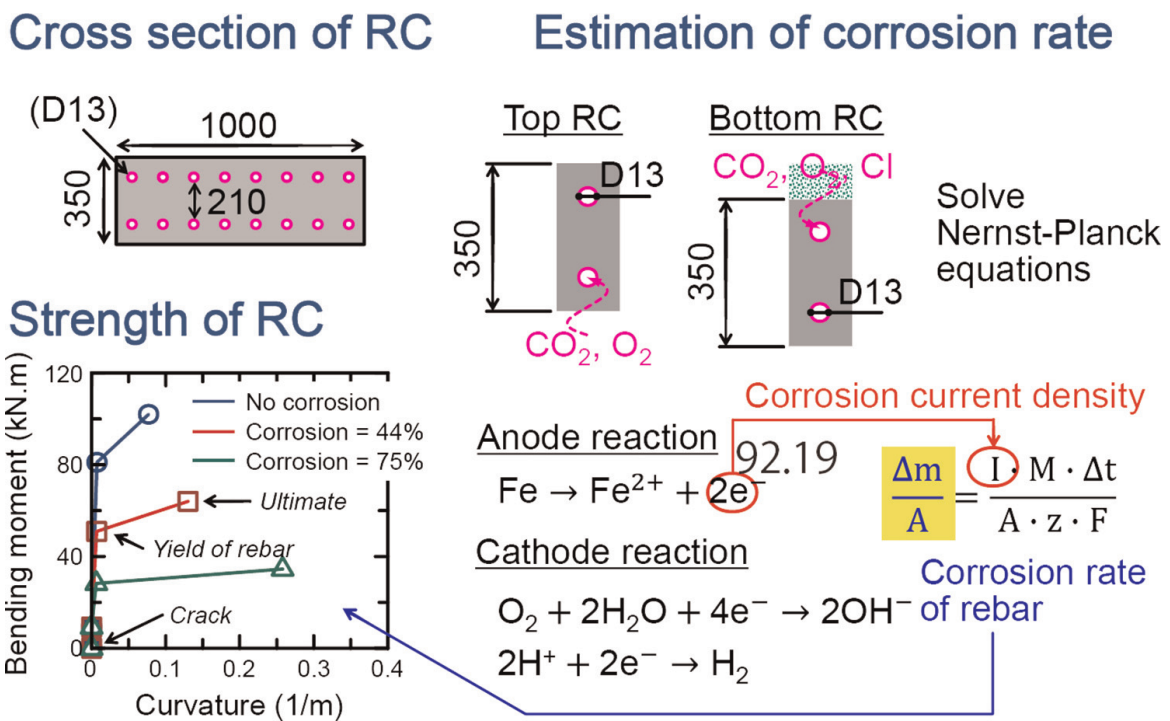


Figure 21. Concept of how to reduce RC strength; from structure analysis given reduced strength, axial force, and bending moment of RC beams are calculated.

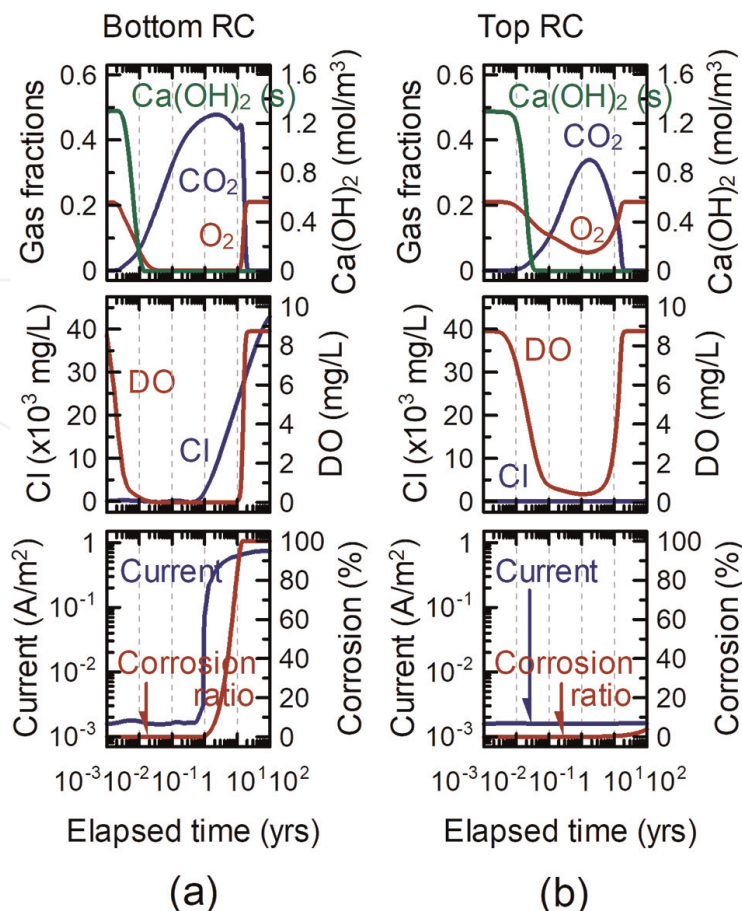


Figure 22.
 Concentration profiles in the containment facility; (a) bottom RC, (b) top RC.

Parameter	Value
N-values for ground assuming sandy loam, N	30
Shear wave velocity, V_s	249 m/s
Maximum oxygen consumption rate, λ_{O_2max}	$4.9 \times 10^{-2} \text{ d}^{-1}$
First-order decay rate in aerobic condition, k^*	2.0 yr^{-1}
First-order decay rate in anaerobic condition, K	0.2 yr^{-1}
Second-order reaction rate for carbonation, λ	$0.6 \text{ m}^3/\text{mol/d}$
Intrinsic permeability for ground, K	$1.0 \times 10^{-12} \text{ m}^2$
Distribution coefficient for sorption layer, K_d	300 mL/g

Table 4.
 Main analytical conditions.

momentum balance to perform ground motion analysis and structure analysis. In the ground motion analysis, a governing equation having an unknown variable of horizontal displacement is solved using a given seismic wave as a boundary condition on the base layer. Then the calculated horizontal displacement and shear stress are applied as external forces acting on the landfill. Finally, a structural analysis for RC-based beams simulating the landfill geometry is conducted. From the calculated axial forces and bending moments in the beams, the crack width is estimated according to previous experimental studies [66–69]. The estimated crack width is used to

		Bending moment (kN.m)	Axial force (kN)	Failure mode	Crack width (mm)	Flow rate (mL/d/m)
Case 1	Bottom RC	6.6	39.9	No crack	0	Close
	Top RC	23.2	11.7	Crack	0.330	2900
Case 2	Bottom RC	6.2	37.9	No crack	0	Close
	Top RC	20.9	17.1	Crack	0.291	1990
Cases 3 and 4	Bottom RC	7.3	25.9	Crack	0.110	Close
	Top RC	22.2	17.9	Crack	0.306	2310

Table 5.
Results of seismic analysis.

determine the leakage rate from inside to outside the landfill according to the theory of Poiseuille flow. The governing equations mentioned above are solved using Moleman-i plus (Mizuho Information & Research Institute) for seismic analysis and COMSOL Multiphysics ver 5.0 for reactive chemical transport analysis.

Table 5 shows the results of the seismic analysis. In any top cover with reinforced concretes, a crack is generated, resulting in the infiltration of rainfall into the landfill compartment. Whereas a crack in the bottom-reinforced concrete is not generated in Cases 1 and 2, deterioration due to carbonation and salt damage is considered. The crack width in Cases 3 and 4 is estimated to be 0.11 mm, which is smaller than that in the concrete reinforced with a top cover consisting of reinforced concrete. **Figure 23** shows the results of groundwater flow and transport analysis for Cd leached from the waste containment facility. No Cd appears in Cases 1 and 2 because the bottom-reinforced concrete does not have a crack. However, Cd does leak from the crack in Cases 3 and 4; it is transported by groundwater flow and observed in the monitoring well. The soil sorption layer is effective for preventing groundwater contamination, and it is an important factor in controlling the long-term environmental safety of isolation-type landfills.

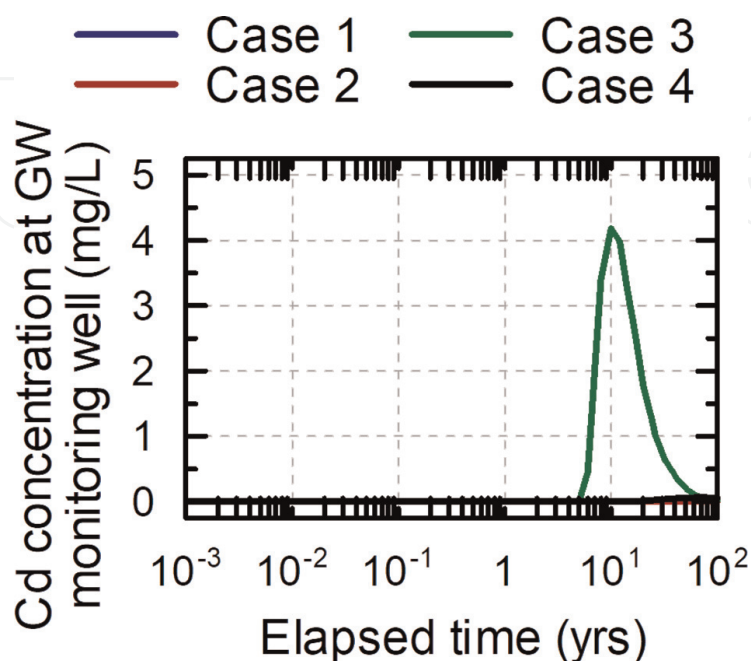


Figure 23.
Cd concentration profiles in groundwater.

This section presents a numerical simulation model to evaluate the environmental safety of hazardous waste landfills having a reinforced concrete structure. A multi-barrier system, in which a sorption layer is additionally installed under a reinforced-concrete hazardous waste landfill, can retard the transport of contaminants and will be effective for improving their environmental safety.

7. Concluding remarks

To develop an environmentally sound resource circulation society, it will be essential to implement long-term safety management practices for hazardous substances. The necessity of isolation/sequestration schemes for hazardous heavy metals is described in this chapter in terms of the multi-barrier concept, in which engineering measures, such as pretreatment and solidification technologies, control of landfill conditions, isolation barriers, and geological and artificial barriers, are implemented to attenuate environmental risks. Single artificial engineering measures should always be viewed with skepticism, and multiple barriers are necessary to retard and attenuate the migration of hazardous heavy metals. The multi-barrier concept also considers the (bio) chemical transformation behavior of heavy metals and their effect on migration. A further innovation of measures to isolate/sequester heavy metals may lead to higher levels of safety and more environmentally sound resource circulation.

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Conflict of interest


The authors declare no conflict of interest.

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