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# Characteristic and remediation of radioactive soil in nuclear facility sites: a critical review

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#### Abstract

A huge amount of radioactive soil has been generated through decommissioning of nuclear facilities around the world. This review focuses on the difficulties and complexities associated with the remediation of radioactive soils at the site level; therefore, laboratory studies were excluded from this review. The problems faced while remediating radioactive soils using techniques based on strategies such as dry separation, soil washing, flotation separation, thermal desorption, electrokinetic remediation, and phytoremediation are discussed, along with appropriate examples. Various factors such as soil type, particle size, the fraction of fine particles, and radionuclide characteristics that strongly influence radioactive soil decontamination processes are highlighted. In this review, we also survey and compare the pool of available technologies currently being used for the remediation of radionuclide-contaminated soils, as well as the economic aspects of soil remediation using different techniques. This review demonstrates the importance of the integrated role of various factors in determining the effectiveness of the radioactive soil decontamination process.

Keywords Decommissioning . Radioactive soil . Radionuclide characteristics . Remediation . Cesium . Disposal

# Introduction

The Fukushima Daiichi nuclear accident in Japan caused severe environmental pollution by radionuclides, contaminating large amounts of soil  $(\sim 28 \text{ million m}^3)$  over a wide area (Yamamoto [2012\)](#page-15-0). The aftermath of the Fukushima Nuclear Power Plant (NPP) accident included an increase in social interest in decommissioning old NPPs and controversy about expanding NPPs. In addition to this critical accident, unintended leaks of radioactive materials from various nuclear facilities have contaminated the environment, and most radionuclide-contaminated soils are considered low-level radioactive waste (Zhu and Shaw [2000\)](#page-15-0).

Leakage during the operation of nuclear facilities can result in radioactive contamination of the soil at a decommissioning

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 $\boxtimes$  In-Ho Yoon [ihyoon@kaeri.re.kr](mailto:ihyoon@kaeri.re.kr) site. A large amount of soil waste contaminated with  $^{137}Cs$ ,  $90$ Sr,  $60$ Co, and  $3$ H has been generated during the decommissioning of international commercial NPPs (Lee et al. [2019\)](#page-15-0). At the Connecticut Yankee NPP (USA), the generated radioactive soil wastes were amounted to 1.17 million  $ft<sup>3</sup>$  (33,000 m<sup>3</sup>) (EPRI [2006\)](#page-14-0). Approximately 30% of all the waste from decommissioning the Maine Yankee NPP (USA) was radioactive soil (EPRI [2005\)](#page-14-0). Details about the contamination of radioactive soil are summarized in Table [1.](#page-1-0) Internationally, environmental projects for site remediation after decommissioning nuclear facilities used for nuclear weapons manufacturing and research are ongoing (Alvarez [2016\)](#page-14-0). Environmental remediation costs have averaged \$60 million per unit for the decommissioning of commercial NPPs in the USA, accounting for 10% of the average decommissioning cost (OECD-NEA [2016\)](#page-15-0). In the case of commercial NPPs, 540 NPPs are expected to be decommissioned by 2047 and projects for the remediation of radionuclide-contaminated environments due to NPP decommissioning are expected to expand dramatically (Volk et al. [2019](#page-15-0)).

Understanding the characteristics and transport phenomena of radionuclides is important for their remediation. Recently, radionuclides such as Pu and Am have been shown to adsorb

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onto soil particles and to transfer in particulate form (Clark et al. [2006;](#page-14-0) Kersting [2013](#page-14-0)). In the process of site characterization, contaminated soil was excavated on the basis of measurements of the degree of its radioactivity level, and the excavated soil was collected in a large tent. Excavation is a good remediation method for radioactive soils; however, the cost of remediation varies greatly depending on the amounts of radioactive soils and the costs are associated with transporting them to a disposal site. If the amount of radioactive waste is substantial, the application of a volume reduction technology is required to reduce disposal costs to below the clearance level. In conjunction with advances in cleanup technologies and increasing disposal costs, extensive research has recently been conducted to reduce the volume of radioactive waste (Agnew et al. [2011](#page-14-0)). Subsequent separation and treatment processes were performed for volume reduction. An evaluation of the costs was required to determine the feasibility of this final process.

An optimal treatment technology is critical for the generated soil waste at decommissioned sites. Factors to be considered when dismantling domestic nuclear facilities in the future include the generation of a potentially large amount of radioactive soil. Although substantial progress has been made in the development of soil remediation techniques for some radionuclides such as U and Co, the removal of 2:1 clay minerals containing radioactive Cs from the soil has remained relatively inefficient or has required high energy consumption processes because of the strong and irreversible interactions (Park et al. [2017\)](#page-15-0). Therefore, studies have been performed to investigate remediation technologies applicable to soils contaminated with radioactive Cs. Existing soil remediation techniques are classified into dry separation, soil washing, floatation separation, thermal desorption, electrokinetic remediation, phytoremediation, and integrated technologies (Fang and Achal [2019](#page-14-0); Hussain et al. [2018](#page-14-0); Kumbhar et al. [2021\)](#page-14-0). The principles and processes of these various techniques are surveyed and critically compared herein.

In this review, we discuss the key soil characteristics and treatments faced by various researchers while treating radioactive soils in actual contaminated sites using conventional remediation methods (excavation, segmentation, and detection). Because laboratory studies have been conducted using various techniques, materials, and strategies with spiked radionuclides and these studies have been extensively reviewed elsewhere, we have excluded laboratory studies from further discussion in this review. In this paper, we first review the characteristics of the radioactive soil to be treated and then focus on trends in the research and development of soil treatment technologies in field tests. Finally, we discuss the limitations of existing technologies for fine particles and consider directions for technical improvements in the volume reduction of radioactive soil waste.

# Characteristics and mobility of radionuclide-contaminated soil

# Characteristics of radionuclide-contaminated soil

The radionuclides listed in Table 2 are those that generate the greatest environmental concern. Radionuclides undergo the same reaction in soil as the non-radioactive isotopes of the element (Wild [1993\)](#page-15-0); however, the concentration of radionuclides in the soil is generally very low compared with the concentrations of non-radioactive isotopes. When radionuclides enter the soil environment, their mobility is established by both internal factors (physicochemical properties of the soil) and external factors (climate and vegetation) (Fig. [1\)](#page-3-0) (Zhu and Shaw [2000\)](#page-15-0).

The interaction between radionuclides and soil involves physical (reversible) sorption controlled by uncompensated surface charges on soil particles and chemical (principally irreversible) sorption through high-affinity specific interactions and the formation of covalent bonds (Sparks [2003](#page-15-0); Sposito [2008\)](#page-15-0). The primary minerals of the soil, mainly quartz and feldspar, originate from the parent rock and compose most of the sand and silt fraction. Sand and silt play a less-pronounced role in radionuclide interactions, and attachment occurs through reversible adsorption due to the relatively low specific surface areas of sand and silt (Yong et al. [1992](#page-15-0)). Secondary minerals (clay) are the result of the physical, chemical, and biological weathering processes of primary minerals. Because of the unbalanced charges of structural ions, they are permanent surface charge carriers that combine with small particles with large specific surface areas to create a matrix that plays an important role in contaminant retention. In addition, the oxides and (oxy) hydroxides of Fe and Al are rich in amorphous forms and have pH-dependent surface charges. Soil organic matter comprises chains of carbon atoms that contain polar and/or ionized surface agents (e.g., OH<sup>T</sup> and COOH<sup>T</sup>). As a result, clay minerals, Fe and Al oxides, and organic materials react and undergo various interactions with contaminants (Yong et al. [1992\)](#page-15-0). In particular, the removal of radioactive Cs contained in 2:1 clay minerals in soil remains relatively inefficient or requires high energy consumption because of the strong and irreversible interactions (Park et al. [2017\)](#page-15-0). Micaceous minerals intercalate cations (e.g., dehydrated  $K^+$  ions) between the layers, where the cations have negative charges. These intercalated cations are not interchangeable, unlike the cations of expanded smectite. Nevertheless, weathering at the edges of the mica mineral has been reported to expand the interlayer space and enable the partial exchange of interlayer cations (Benedicto et al.  $2014$ ; Sawhney [1972\)](#page-15-0). Weakly hydrated  $Cs<sup>+</sup> ions$ adsorbed onto the expanded interlayers of the peripheral area around the mica mineral have been reported to undergo dehydration and induce the partial collapse of the interlayers (Fig. [2\)](#page-3-0). This structural collapse makes subsequent desorption of other cations difficult (Nakao et al. [2008;](#page-15-0) Sawhney [1972](#page-15-0)).

#### Factors affecting the mobility of radionuclides in soil

A soil's ability to immobilize radionuclides is the main factor controlling the bioavailability of such radioactivity, and this factor works in conjunction with numerous external factors. The structure and size of the soil, mineral components, organic components, redox potential (Eh), and pH, as well as rainfall, climate change, and soil management, affect the mobility of radionuclides (Iurian et al. [2015](#page-14-0)). Soil pH, cation exchange capacity (CEC), and total organic carbon (TOC) are the physical and chemical properties most often associated with the distribution of radionuclides (Smičiklas et al. [2015\)](#page-15-0). The structure of minerals is a major factor affecting soil CEC because it governs the soil's ability to retain important nutrients and contaminants. The particle size of soil is based on its relative fractions of sand (particle size  $50-2000 \mu m$ ), silt (2– 50  $\mu$ m), and clay (<2  $\mu$ m) (Sparks [2003\)](#page-15-0). Of these components, clay fractions have the largest specific surface area because of their very fine particles and play an important role in chemistry and CEC. However, they also exhibit an important holding capacity for transporting nutrients and pollutants to



Table 2 Characteristics of n radionuclides in soil (Zhu and Shaw [2000](#page-15-0))

<span id="page-3-0"></span>

soil organisms and plants. In addition, soil organic matter contributes substantially to the soil CEC and to the soil moisture retention capacity.

# Remediation technology of radionuclide-contaminated soil

### Excavation

Excavation is commonly used in soil remediation work because it is a very simple and effective remediation technology for both surface and subsurface soil. Soil replacement of



Partial collapse of the interlayers

Fig. 2 Schematic of Cs adsorption and transport in clay minerals

contaminated soil refers to replacing or partly replacing contaminated soil with noncontaminated soil. The soil replacement method dilutes the concentration of radionuclides in soil, which in turn increases soil functionality (Yao et al. [2012\)](#page-15-0). The replaced soil is generally treated to remove radionuclides or, in some cases, dumped into the final disposal site. Soil replacement methods can effectively isolate contaminated soil and ecosystems, and minimize the environmental impact of radionuclides. However, because soil replacement is laborintensive and therefore expensive, it is most suitable for soil with high pollution levels and a small area. Bulk excavation, short-distance transportation, and disposal costs range from \$270 to \$460 per ton (Khalid et al. [2017\)](#page-14-0). Long-distance transportation of excavated soil can dramatically increase the cost. In addition, this technology may not be applicable to agricultural sites because it introduces a risk of soil fertility loss. Extensive excavation has been performed over a wide area at every decommissioned NPP site to reduce residual activity by removing contaminants. Special excavation techniques have also been used in specific areas on a case-by-case basis (see Table [1\)](#page-1-0).

### Volume reduction

#### Separation

Dry separation Dry separation is one of the most widely used techniques for treating radioactive soil contamination. This technology is used to separate contaminated and noncontaminated soil in dry conditions using a real-time radio detector while excavated soil is transported on conveyor belts (Bayliss and Langley [2003;](#page-14-0) EPA [2007\)](#page-14-0). Large rocks or other

objects (e.g., plants and roots) are first removed from the excavated soil by screening, and the soil is subsequently placed onto a conveyor belt for the next process. Rocks and gravel can be sorted by dry separation after being homogenized through a crushing process. Separated noncontaminated soil is used for backfill, and contaminated soil is disposed of as radioactive waste. Dry separation is one of the most widely used techniques in the treatment of radioactive-contaminated soil. This dryseparation approach can be used to treat large amounts of soil quickly and to separate the soil; it can also be used to treat contaminated materials such as concrete and asphalt. However, in field tests, the reduction efficiency of radioactive soil waste has been found to strongly depend on the characteristics of the contaminated soil. In some cases, low separation efficiency resulted from the relatively homogeneous distribution of radioactive contamination and low content of noncontaminated soil because the contaminated area was only partially excavated. In addition, a device is required for controlling the dust generated during the treatment process. The separators also become waste after the separation process is completed. Therefore, the characteristics of contaminated soils should be analyzed through preliminary experiments to assess the applicability of dry separation techniques (Giles [1999](#page-14-0)).

In general, gamma scintillation detectors, such as those based on NaI, can be used to separate soil contaminated with gammaemitting radionuclides, and alpha- and beta-emitting contaminants can also be predicted using scale factors. Therefore, gamma detectors can be used to separate soil contaminated with gamma nuclides (e.g.,  $^{232}$ Th,  $^{238}$ U,  $^{137}$ Cs, and  $^{60}$ Co). In addition, researchers at Sandia National Laboratories (USA) have reported using beta detectors to separate soil contaminated with  $\frac{90}{9}$ Sr (among other radionuclides) (DOE [1999\)](#page-14-0). In particular, the dry-separation method can be applied to soil that is heterogeneously contaminated with excavated soil; however, it is less applicable to homogeneously contaminated soil (DOE [1999\)](#page-14-0).

In field applications, commercial dry separators such as the Segmented Gate System of Eberlin Services (formerly Thermo Nutech) have been developed for the treatment of 239Pu-contaminated soil at an early US military base (the Johnston Atoll DoD site) (DOE [2001;](#page-14-0) Rogers [1998\)](#page-15-0). More than  $70,000 \text{ m}^3$  of excavated soil was treated, and a high rate of volume reduction (greater than 90%) was achieved (Cummings and Booth [1996;](#page-14-0) EPA [2007;](#page-14-0) Moroney et al. [1994\)](#page-15-0). Because applicability assessments have been conducted at various EPA and DOE sites in the US, the performance of dry separation technology can be summarized as shown in Table 3(EPA [2007](#page-14-0)). Researchers at the Idaho National Engineering and Environmental Lab (INEEL) attempted to separate contaminated soil using Thermo Nutech's Segmented Gate System for the treatment of <sup>137</sup>Cs-contaminated soil generated at site ARA-23 (USA). The contamination standard for separation was 23 pCi/g and a total of 338 m<sup>3</sup> of soil was injected. However, the results showed that dry separation technology was not applicable because the volume reduction rate was less than 3%. This result suggested that the contamination standard should be 110 pCi/g to reduce the volume by more than 90%. The low separation efficiency was the result of a relatively homogeneous distribution of radioactive contamination and low content of noncontaminated soil because the contaminated area was only partially excavated. Therefore, the characteristics of contaminated soils should be analyzed for their applicability to dry separation techniques through preliminary experiments (Giles [1999\)](#page-14-0).

Wet separation Wet particle-size separation is generally conducted in conjunction with soil washing because radionuclides exhibit the greatest affinity for particles with very high specific surface area/volume ratios, such as silts and clays. During wet separation with washing, the scrubbing action

Table 3 Volume reduction by dry separation technology (EPA 2007)

<b>Site</b>	Radionuclides	Separation criteria $(pCi/g)$	treated $(m^3)$	Amount of soil Volume reduction
Johnston Atoll	Pu-239, Am-241, Ra-222 13		76.453	Maximum $99.5\%$
Los Alamos National Laboratory	$U-238$	50	1931	91.6%
Pantex Plant	$U-238$	50	225	38.5%
Sandia National Laboratories, Site 16	$U-238$	54	506	99.9%
Sandia National Laboratories, Site 228A	$U-238$	27	1034	99.5%
Tonapah Test Range, Nevada	$Pu-239$	50-1500	255	Maximum 99%
Idaho National Engineering and Environmental Laboratory Cs-137		23	338	$<3\%$
Brookhaven National Laboratory	$Cs-137$	23	478	$16\%$
New Brunswick FUSRAP	U-238, Th-232, Ra-226	5	3823	55%
Savannah River Site	$Cs-137$	4	917	99%
West Valley Nuclear, New York	$Cs-137, Sr-90$	45	460	61%

generally removes surface contaminants from large soil particles such as sand and gravel. The fine soil particles can be further separated in a sedimentation tank, sometimes with the aid of a flocculating agent. The output streams of the soil washing process include clean granular soil particles, contaminated fine soil particles, and washing solution (Fig. 3).

Soil decontamination was carried out by aqueous scrubbing at the Spain Cabrera NPP. Initially, the contaminated soils from the excavation were continuously processed at the plant, where the fine fraction (clays and silts), which was the most contaminated, was separated from the coarser fractions (sand and gravel), which were removable in general. The coarse fractions were then subjected to a physical washing process and finally managed as declassifiable material. The goal was conventional management once radiological determinations were conducted to ensure compliance with the declassification level. The contaminants concentrated in the fine fraction then represented a small volume that was managed as low-activity radioactive waste. In this way, the total volume of radioactive waste was reduced, with a consequent reduction in the cost associated with managing the affected soils during remediation of the site (Correa Sáinz and Martín Palomo [2020\)](#page-14-0).

Flotation separation In flotation separation, contaminated fine particles from total soil are separated using a technique based on the surface properties of the particles. A foam-friendly material is attached to the surface of foam by injecting gas into the dispersion solution or slurry containing fine particles from contaminated soils. The particles attached to the foam

Fig. 3 Schematic of a soil washing process

are then floated together with the foam and subsequently separated from the upper part. In general, hydrophobic particles with a low affinity for water attach to the gas-liquid surface of the foams and then float with the foam. By contrast, hydrophilic particles sink into the water. These floating separations are affected by the surface charge, particle size, and other properties of the fine particles.

To increase the efficiency of floating separation, the targeted fine particles containing radionuclides can be optionally changed to a hydrophobic form by adding floating agents such as surfactants. The surfactants physically and chemically adsorb onto the surface of a contaminated particle, resulting in a further increase of its hydrophobicity. In general, surfactants with an ionic head group and hydrophobic chains are used and are attached to charged particles. The mixture of surfactant and fine particles results in hydrophobic chains exposed at the particles' surface, which in turn results in a increase of the hydrophobicity of the particles. When the concentration of surfactant increases, the formation of a double layer by hydrophobic interaction with the surfactant may reduce the hydrophobicity, resulting in a decrease in the flotation efficiency. Therefore, the concentration of the surfactant must be properly controlled to increase the flotation efficiency. In addition, the interaction between the surfactant and fine particles can vary depending on the pH of the solution. Foaming agents are also added to accelerate the formation of foams for floating selection.

Floating separation, which is widely used in the smelting process, has also been evaluated for its applicability to the remediation of radioactively contaminated soil containing



radionuclides such as U, Ra, and Cs. Flotation separation is a technique for separating contaminated fine particles from mixed soil and is based on the surface properties of particles. Additives such as oil and hydrophobic aggregates are needed to improve the separation efficiency. However, surfactantcontaining wastewater is generated after the process and is difficult to treat.

In field applications, floating separation techniques have been researched to separate U from Elliott Lake Ore in Canada (Muthuswami et al. [1983;](#page-15-0) USEPA [1988\)](#page-15-0). A floating separation technique was applied to remove Ra-226 contamination at the laboratory level from Ottawa soil in Illinois (USA)(Misra et al. [2001\)](#page-15-0). Soil analysis at the Ottawa site showed that Ra-226–contaminated soil was combined with sulfates and exhibited very low solubility. The floating separation technique reduced the volume by more than 80% and decreased the contamination level to  $<6$  pCi/g.

The CEA (Commissariat à l'énergie atomique et aux énergies alternatives, translated as the French Alternative Energies and Atomic Energy Commission) investigated the separation of anionic clay components extensively contaminated with radioactive Cs, using a cationic surfactant. The cationic surfactant was combined with illite clay minerals to provide a negative charge to render the surface of the clay minerals hydrophobic. After the clay minerals interacted with the cationic surfactant, they were sorted using flotation separation techniques (Chapelain et al. [2016](#page-14-0); Zhang et al. [2019](#page-15-0)).

In addition, the CEA used the floating screening technology developed as a floating separation technology for soil remediation via the removal of clay minerals from highly Cscontaminated soil in Fukushima, Japan. In 2017, the Japanese government's "Decontamination technology demonstration test project" also demonstrated the treatment of contaminated soil from Fukushima, Japan. Approximately 70–85% of the total amount of soil was separated from the Cs-contaminated soils, and Cs contamination of the separated soils was reduced to 33–50% (JAEA [2018\)](#page-14-0).

Magnetic separation Magnetic separation is a technique for reducing the radioactivity of contaminated soil by separating clay particles that contain a high Cs concentration. Various methods have been studied to reduce the volume of radioactive soil. Magnetic separation, in particular, has attracted attention because it is a physical method that does not use adsorbents or chemical reagents (e.g., acid, cations, chelating agents, or surfactants). Other advantages of magnetic separation include simple operation, low energy consumption, and low cost.

In field applications, extensive research has been conducted to separate fine particles with a high concentration of radioactive Cs from the actual Fukushima soil using modified  $Fe<sub>3</sub>O<sub>4</sub>$  particles under dry conditions. Fine soil particles coated with magnetic particles are attracted to a magnet, even against gravity, and the waste is produced in a dry state that is easy to store. Thus, this method can reduce the volume of the contaminated soil by concentrating radioactive Cs into a smaller volume (Sato et al. [2019\)](#page-15-0).

### **Treatment**

Soil washing Soil washing is one of the most common industrial treatments for non-radioactively contaminated soils, and the soil washing process has been used to treat soils contaminated with U, Ra, Th, Sr, and Cs (Devgun et al. [1993](#page-14-0); EPA [1990](#page-14-0)). The soil washing technique includes pre-treatment, washing, separation, and wastewater treatment processes. The order and number of cleaning steps of the washing process are determined by the characteristics of the contaminated soil. In general, large objects such as branches and gravel from soil excavated during pretreatment are physically removed by scalping and mechanical screening. As a result, relatively homogeneous soil is obtained. In addition, a process for dispersing soil-forming clusters is also carried out to enhance the cleaning effect. The effectiveness of the soil washing process is determined by the properties of both the contaminants and the soil components. Soil washing is effective only if radionuclides can be readily desorbed from large soil fractions, and if the contaminated soils (optimally) contain less than 25% silt and clays. To improve the removal efficiency of contaminants from soils, surfactants, acids, or solvents can be added to the washing solutions (Hubler and Metz [2013](#page-14-0)).

Soil washing is quick and extensive; however, there are specific requirements depending on the soil texture. Sandy soils with high porosity, good permeability, and adequate moisture content exhibit a better effect from soil washing, whereas soil washing is not as effective for clay soils or soils with high moisture content because of their low porosity and low permeability and the dilution effect of soil water. During soil washing, the contaminated soil is mixed with suitable extractants depending on the type of soil and radionuclides. The extract and soil are thoroughly mixed for a certain period. Precipitation, ion exchange, chelation, or adsorption is used to transfer radionuclides from the soil to a liquid phase and to separate them from the leachate (Ferraro et al. [2015](#page-14-0)). For compliance with regulatory criteria, the separated soil can be returned to and buried in its original location. Soil washing is also a quick method to satisfy certain criteria without assuming long-term responsibility (Park and Son [2017](#page-15-0)). Because of its high efficiency, soil washing is considered one of the most cost-effective soil remediation technologies. The reagents used in soil cleaning are selected on a case-by-case basis, and their use and efficiency depend on the type and location of the radionuclides. The exchange/extraction/availability of radionuclides during soil washing is known to depend on the soil and the type of radionuclide (Liao et al. [2016](#page-15-0)). The efficiency of soil cleaning depends on the ability of the extractor

to dissolve radionuclides in the soil. Therefore, extractants that can dissolve high concentrations of metals are suitable for soil washing.

In field applications, the soil washing technique was initially evaluated for applicability to contaminated soil at the Rocky Flats site in the USA. The soil was contaminated with Am and Pu at 900–140,000 pCi/g. Most of the contaminated soil was composed of large particles such as gravel. After wet soil separation, the level of contamination was less than 3 pCi/ g in the soil and the percentage of particles larger than 2 mm was reduced to 65%. The contamination level of soil particles from 2.4 to 0.42 mm was reduced to 300 pCi/g through wet soil separation. Moreover, using friction scrubbing, researchers further reduced the contamination level to <20 pCi/g(Karthika et al. [2016;](#page-14-0) Navratil and Kochen [1982](#page-15-0)). The washing efficiencies of <sup>239</sup>Pu and Am-241 contaminated soils, which were obtained from five DOE sites (i.e., Rocky Flats, Hanford, Mound, Idaho National Engineering Laboratory (INEL), and Los Alamos National Laboratory (LANL) in the USA), were evaluated on a bench scale. The washing solutions were evaluated using a NaOH solution (pH 12.5),  $2\%$  HNO<sub>3</sub> solution containing 0.2% HF solution, and a 2 N HCl solution. The NaOH solution was only successful for the Rocky Flat soils, where it reduced the concentration of Pu to 2.5% in soils with a particle size greater than 105  $\mu$ m. The HCl solution showed good washing efficiency for the soils at Hanford, INEL, and Mound. After the Hanford soil (>105 μm) was washed with HCl, the concentrations of Pu and Am decreased to 4.4 and 1%, respectively (Stevens and Rutherford [1982\)](#page-15-0).

In Japan, Konoike E&C evaluated the applicability of a jet cleaning process by crushing and washing gravel (5 mm or larger) and soil  $(<5$  mm) contaminated with 10 Bq/g or more

Fig. 4 Thermal-based soil remediation process for Cscontaminated soils

of Cs. After the sand or sandy soil (75 μm or larger) was separated and the moisture was removed using a filter press, the volume reduction rate was ~70%. At Sato Kogyo Co., the soil was washed with high-pressure water, followed by the separation of the soil particles smaller than 300 μm. In addition, the applicability of a process for removing soil with a particle size 75–300 μm via a washing step using microbubbles was evaluated for soil contaminated with  $6-7Bq/g$  of Cs. Approximately 88% of the decontamination effect was achieved through soil washing.

Thermal desorption Thermal desorption is a technique used to increase Cs desorption by exposing contaminated soil to high temperatures and inducing vaporization or interlayer expansion through pyrolysis (Fig. 4). In addition, it increases the efficiency by increasing the activity of the desorbent and by expanding the interlayers between contaminated soils because of the high temperature. Although thermal desorption is effective for purifying Cs-contaminated soil, it requires a large amount of energy compared with other treatment techniques and thus has a high treatment cost. Therefore, thermal desorption is better suited to maximizing the effect of other treatments rather than being used as a standalone treatment. In thermal desorption, Cs desorption is increased by exposing contaminated soil to high temperatures and inducing vaporization or interlayer expansion through pyrolysis. It is effective for volatile radionuclides such as Cs. However, it requires a large amount of energy compared with other treatment techniques and has a high treatment cost.

In a field application study at Oak Ridge National Laboratory (ORNL), only  $137Cs$  was observed to be substantially more vaporized than expected during a process conducted to stabilize radioactively contaminated soil using



underground vitrification. Subsequently, thermal desorption research was conducted to exploit the volatility of Cs at high temperatures. ORNL researchers observed thermal desorption of Cs from contaminated soil at high temperatures (700–1300 °C) and added various additives to improve the extent of thermal desorption of Cs (Shimoyama et al. [2014\)](#page-15-0). The addition of NaCl was the most efficient treatment for removing Cs, and it was shown that more than 99% of  $137Cs$  could be removed at 1000 °C. However, this was not effective after the addition of carbonate, nitrate, phosphate, and sulfate. During the heat treatment process, the clay mineral structure containing Cs collapsed, and a reaction between Cs and Cl was accelerated for the vaporized Cs.

The Japan Atomic Energy Agency (JAEA) studied the conditions necessary to remove Cs by thermal desorption of clay minerals to purify soil contaminated with Cs after the Fukushima accident (Honda et al. [2017a;](#page-14-0) Honda et al. [2017b](#page-14-0); Shimoyama et al.  $2014$ ). When NaCl–CaCl<sub>2</sub> was used as an additive, Cs, which had been combined with clay minerals at 700 °C, was found to be converted to CsCl. Further experiments were conducted to reduce the heat-treatment temperature through decompression and cleaning. Taiheiyo Cement of Japan evaluated the thermal desorption process for decontaminating Cs-contaminated soil at the pilot scale. Heat desorption was induced by heating soil contaminated with 5– 7Bq/g to 1300  $\degree$ C inside a rotary heating system, and the Cs in the exhaust system were removed using a filter. After purification, the contamination level of the soil was found to be less than 0.05 Bq/g, indicating excellent purification of the Cscontaminated soil. However, high energy costs due to the relatively slow processing speeds and high processing temperatures were shown to be problematic.

Electrokinetic remediation Electrokinetic (EK) remediation is an electrochemical extraction process that enables the separation and extraction of radionuclides from saturated or unsaturated soils. It is performed by applying a low voltage direct current to a pair of electrodes on each side of a contaminated soil mass. This current mobilizes ions and charged compounds so that they migrate toward the electrodes. Dissolved ionic species, including radionuclides, migrate to oppositely charged electrodes at a rate that depends on the local potential gradient (this transport mechanism is known as electromigration). Moreover, the extraction of metals from soil is enhanced through the application of acidic conditions around the anode and by the movement of the pore fluid under a potential difference (i.e., electroosmosis) (Fig. 5).

In general, radionuclides such as U, Cs, and Co are cations; these radionuclides, therefore, migrate toward the cathode. With current moving from an acid pole at the anode to a base pole at the cathode (and back), the resultant acid conditions improve the radionuclide mobility and facilitate the removal of radionuclides from the cathode. The main advantage of the EK technique is that it enables in situ remediation without excavation of the soil. This in situ process essentially reduces the costs associated with secondary waste generation, transportation processes, soil excavation, transportation, and disposal. However, the effectiveness is diminished in soils with a moisture content of less than 10%, which adversely affects the soil's electrical conductivity. (EPA [2002;](#page-14-0) Jung et al. [2015\)](#page-14-0). The removal of radionuclides such as Cs present in lowconductivity forms such as sulfide or metallic forms requires preliminary dissolution. In such cases, the use of appropriate electrolytes (e.g., distilled water, organic acid, inorganic acid, or synthetic chelate) can improve the removal efficiency of the





electrokinetic remediation method (Iannelli et al. [2015](#page-14-0)). However, the removal efficiency depends on the type of chemical (anolyte) and the metal recovery method used (Vocciante et al. [2016](#page-15-0)).

In a field application, Sandia National Laboratories (SNL) in the USA used the EK technique to remove uranium from soil. The concentration of the U contaminating the soil was 4000 ppm. The removal efficiency was ~75–95% (Booher et al. [1997](#page-14-0)). However, the EK technique produced nonsoluble deposits when applied to Ra- and Th-contaminated soil, resulting in lower removal efficiency. The Pacific Northwest National Laboratory (PNNL) in the US fabricated a benchscale EK device for the treatment of Cs-contaminated soil. After soil deposits (76% sand and 24% silt) were artificially contaminated with Cs at the Hanford site, electrokinetic remediation was applied for  $~68$  days. The Cs removal efficiency was ~47%, and the removal efficiency in silt and clay components was higher than that in the sand (Jung et al. [2015](#page-14-0)).

Phytoremediation Phytoremediation is the process of using plants to manage contaminants in soil. Phytoremediation mechanisms applicable to solid media include plant extraction, plant degradation, and plant stabilization. Because radionuclides such as Cs are not biodegradable, the main mechanism applicable to the treatment of radionuclides is plant phytoextraction. Phytoextraction is the absorption of pollutants by plant roots and the accumulation of pollutants in plant shoots and leaves. The plants are then harvested, dried, and discarded in the growing area (Fig. 6). Phytoextraction has been pilot-tested at Brookhaven National Laboratory to remove low levels of Cs and Sr from the soil. Phytoextraction was also tested for the removal of Cs from contaminated soils at Argonne National Laboratory West in Idaho, USA (Lee [2000\)](#page-15-0). After the Fukushima accident in Japan, extraction of Cs from contaminated soil using plants again became an active research topic; however, most studies have reported a Cs removal efficiency of less than 0.3%. This removal efficiency

is insufficient for the removal of radionuclides that strongly bind to soil, such as Cs (Ogata et al. [2015;](#page-15-0) Tamaoki et al. [2016\)](#page-15-0). Phytoextraction is only suitable for soil containing radioactive nuclides that are weakly bound to the soil. Some heavy metals (and metalloids), such as Cr and Pb, are highly immobile in the soil, and their plant extraction is often limited. The efficiency of plant extraction is limited by the effective depth (typically 30 to 90 cm) for plant root growth. However, the application of chelating agents can increase the ability of plant extracts to recover from metal contamination because of the bioavailability of radionuclides in soil because the accumulation and translocation of plants increases.

In field applications, phytoextraction efficiency was evaluated for the removal of radioactive materials by plants after  $137$ Cs and  $90$ Sr solutions were injected directly into the soil where plants were cultivated in a greenhouse. By evaluating the removal efficiencies of various plants, researchers demonstrated that as much as 71% of the Cs and 88% of the Sr could be removed by plants (Entry et al. [1999](#page-14-0)). Meanwhile, phytoextraction was applied to contaminated soil exposed to Cs for 10 years after the Chernobyl accident. To increase the desorption of Cs from contaminated soil, the concentration of the Cs extracted by plants was analyzed after the addition of cationic substances such as ammonium and potassium. Only 10–25% of the Cs from the contaminated soil at Chernobyl could be recovered by plant remediation (Dushenkov et al. [1999\)](#page-14-0). After reacting with soil, Cs bind to create a very stable form over time; the extraction efficiency by the plants is therefore low, and phytoextraction has been found to be unsuitable for the remediation of long-term soil contamination. After the Fukushima accident in Japan, Cs extraction in contaminated soil using plants was again utilized; however, most of the related studies have reported a Cs removal efficiency of less than 0.3%, which is insufficient for the removal of radionuclides that strongly bind to soil, such as Cs (Ogata et al. [2015](#page-15-0), Tamaoki et al. [2016](#page-15-0)). Table [4](#page-10-0) shows the procedure, advantages, and





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 $\frac{1}{2}$ bench, lab scale) and is promising

<sup>2</sup> Treatment process: 000 - Stand-alone technology; 00 - relatively simple (two-process or so); 0 - complex (more technologies, media to be treated, generates excessive waste, etc.) Treatment process: ○○○ - Stand-alone technology; ○○ - relatively simple (two-process or so); ○ - complex (more technologies, media to be treated, generates excessive waste, etc.)

<sup>3</sup> Operation and maintenance intensive: 000 - low degree; 00 - average degree; 0 - high degree Operation and maintenance intensive: ○○○ - low degree; ○○ - average degree; ○ - high degree

4 Capital (capital intensive): 000 - low degree of capital investment; 00 - average degree of capital investment algebre of capital investment 4Capital (capital intensive): ○○○ - low degree of capital investment; ○○ - average degree of capital investment; ○ - high degree of capital investment

<sup>5</sup> System reliability and maintainability: 000 - high reliability and low maintenance; 00 - average reliability and average maintenance; 0 - low reliability and high maintenance System reliability and maintainability: ○○○ - high reliability and low maintenance; ○○ - average reliability and average maintenance; ○ - low reliability and high maintenance

<sup>6</sup> Relative costs: 000-low degree of general costs relative to other options; 00-average degree of general costs of general costs relative to other options; 0-depends  $6$  Relative costs:  $0$   $0$  - low degree of general costs relative to other options;  $0$  - average degree of general costs relative to other options;  $0$  - high degree of general costs relative to other options;  $0$  - dep on scale and operation on scale and operation

Time: 000 - comparatively very less; 00 - average; 0 - more to medium Time:  $000 - \text{comparatively very less}$ ;  $00 - \text{average}$ ;  $0 - \text{more to medium}$ 

disadvantages of soil remediation techniques (Agnew et al. [2011](#page-14-0); Khalid et al. [2017;](#page-14-0) Orr et al. [2014](#page-15-0); Regulation [2020](#page-15-0), USEPA [2007](#page-14-0)).

# Critical challenges faced during the remediation of radioactive soil

# Cost analysis for excavation and treatment (volume reduction)

The treatment (volume reduction) represents an additional process and is complicated when applied to multiple processes. However, it can reduce the amount of radioactively contaminated soils at nuclear facilities. Consequently, an economic assessment of the costs of volume reduction is important because it determines whether volume reduction should be carried out. The cost of excavation and disposal has three main factors: First, decontamination underground can be carried out using various methods ranging from simple drilling of soil to reduce the size for pickup after rock blasting. For the removal of approximately 1.2 million  $ft^3$  of soil during the decommissioning of the Connecticut Yankee NPP, the cost of removing and packing soil was approximately \$17/ ft<sup>3</sup>(Bushart et al. [2006](#page-14-0)). Second, the cost of transporting contaminated soils depends heavily on the transport distance and the mode of transport to the disposal site. Remediation operations at decommissioned sites have shown that transporting large quantities of soil or rock by transferring wastes to local transshipment facilities to be placed on trains or nonshipment vehicles directly from the site is inexpensive. The concentration of radionuclides in soil and rock recovery waste was relatively low at the decommissioned NPP sites. Finally, the costs of waste disposal are site specific. In some cases, lower unit prices can be obtained by reducing the amount of soil through treatment at the decommissioned sites. Each site should use site-specific cost analysis in evaluating the suitability of remediation.

Agnew et al. ([2011](#page-14-0)) have reported that the overall cost of materials, installation, implementation, and decommissioning for field testing (excluding the original cost of the containment unit and staff costs) was £4000, or approximately £1700/m<sup>3</sup> (£1000/ton) of the treated material. By comparison, the approximated direct disposal cost was  $£5000/m<sup>3</sup>$ , which was based on the current disposal costs at the low-level-waste repository facility at Cumbria, UK, and the cost of packaging, verification, transport, etc. (Agnew et al. [2011](#page-14-0)). If large amounts of contaminated soils were generated from NPP and the cost of disposal was higher than the remediation budget, it needed to consider the treatment (volume reduction) for reducing the cost of disposal.

#### Characteristics of each technology

The technical characteristics, applicability, and economic characteristics of each technology are summarized in Table [4](#page-10-0). Several important factors can affect the selection and application of available soil remediation techniques. These factors include advantages, limitations, development status, treatment process, and relative overall cost and performance of different remediation techniques (Table [4\)](#page-10-0). In addition to social and environmental acceptability, the associated cost is an important factor in determining the success and practicality of remediation technologies under field conditions. Few studies have been conducted on the economic aspects of radionuclide-contaminated soil remediation. In general, the contaminated areas (such as the type and depth of soil, the purpose of the recovered area, the depth of groundwater, etc.), radionuclides (e.g., the radionuclide type and its concentration in soil), and remediation technologies have different requirements and work costs.

Physical removal methods typically require large amounts of human and physical resources. Excavation is typically the most expensive remediation method if large amounts of soil need to be removed or disposed of. Chemical remediation (immobilization and soil washing) is a cost-effective method compared with physical remediation methods. The amount of chemicals required in soil washing to remove radionuclides is generally small because comparatively small amounts of chemical extractants such as organic, inorganic acid, and chelating agents are used. However, the cost of soil washing varies with the type of radionuclides and soil as well as with the type of chemical extractants (Khalid et al. [2017](#page-14-0)). For example, soil washing is easier for Sr or Co removal than for Cs removal because of the immobile nature and high affinity of Cs for soil components. Similarly, soil washing requires more chemicals for clayey soil than for sandy soils because of the strong binding of radionuclides in clayey soils. In the case of chemical extractants, the levels/concentrations of synthetic chelates such as ethylenediaminetetraacetic acid (EDTA) or inorganic/organic acid are 10–50 times higher than those of natural ligands such as low molecular weight organic acids (LMWOAs) or humic substances (Shahid et al. [2012](#page-15-0)).

Phytoremediation is relatively economical compared with physical and chemical recovery technologies. All types of phytoremediation generally harness natural processes and treat the radioactive contaminated sites in place, with no excavation or physical removal, thereby reducing the cost of site remediation. In addition, in some cases, phytoremediation can remediate the radionuclides without any human involvement; this natural attenuation results in considerable cost savings. For example, because phytoremediation can be carried out with minimal maintenance after its establishment in the field, phytoremediation costs are almost tenfold less

than the costs of engineering-based methods (Marques et al. [2009\)](#page-15-0). Likewise, post-cleanup costs are very low for phytoremediation methods because these methods cause minimum site disturbance compared with conventional physical cleanup methods.

### Selection of soil remediation technology in sitespecific cases

The remediation plan in site-specific cases can be set up as a small area of high concentration, an intermediate area of intermediate concentration, or a large area of low concentration. The important point to be addressed before the remediation is whether treatment at the removal site (i.e., volume reduction) is necessary. If a small area is contaminated with high concentrations of radionuclides, it can be selected for excavation and disposal rather than using treatment facilities. In the second scenario, if the intermediate area is contaminated with intermediate concentrations of radionuclides, the disposal cost can be reduced by reducing the amount of soil waste treated by treatment facilities. However, an economic assessment should be conducted. In addition, an evaluation of whether soil waste is removable and the most efficient removal method among various technologies is important.

The remediation of radioactive soils varies most depending on environmental conditions but also depends on location. Therefore, before choosing a remediation technique for treating a specific soil, we need to understand the factors that affect remediation and design a strategy appropriate for a specific site. These technologies individually face multifactorial problems; an integrated approach is therefore needed to remediate radioactive soils. For example, soil washing might be conducted for radioactive soil waste, followed by advanced separation and treatment of fine particles using flotation separation and thermal desorption, respectively. Numerous researchers have recently developed techniques to selectively separate and dispose of fine soil, including clay, during the removal of contaminated soil. Hybrid complex processes that simultaneously separate fine soil and remove radionuclides can ultimately reduce the volume of radioactive soil waste by more than 95% (Kim et al. [2020;](#page-14-0) Kim et al. [2021](#page-14-0)). If a large area is contaminated with low concentrations of radionuclides, a biological method (phytoremediation) can be considered. Phytoremediation can be also used for postremediation after the excavation and treatment for a long-term period. In the case of the nuclear accident at Fukushima, the soil was collected in wide areas to a depth of 5 cm and transported to a disposal site before phytoremediation was applied in wide areas.

### Conclusions and perspectives

In this paper, we discuss the technology status of remediation of nuclear reactor sites and the future prospects of this technology. Research to reduce the volume of contaminated soil waste has been conducted for a large amount of soil generated by operational leaks at nuclear facilities, and no substantial geographical or economic difficulty in disposing of the radioactively contaminated soil has been identified. Governments have recently been considering efficient treatment methods through economic evaluations because of increasing disposal and transportation costs. The treatment of radioactively contaminated soil around the world has been an active research topic since the Fukushima nuclear accident; however, no effective remediation methods have been proposed to address this problem.

Although various technologies have been successfully tested in the laboratory and in the field, when used alone, these technologies encounter problems that are multifactorial in nature. Therefore, an integrated approach is needed to remediate radioactive soils. The factors that influence radioactive soil remediation vary according to soil properties such as soil type, particle size, the fraction of fine particles, and the radionuclide characteristics. The presence of fine particles plays an important role in determining the effectiveness of the strategies used, especially in the case of Cs contamination. Radioactive soil remediation mostly depends on environmental conditions and varies from one location to another. Therefore, before a remediation technique is selected for treating a specific soil, we need to understand the factors that affect remediation and design a strategy appropriate for the specific site.

In the future, the radioactively contaminated soil generated during decommissioning of nuclear facilities will be remediated after a remediation strategy based on an efficient cost analysis has been established. In addition, many researchers have developed effective remediation methods for radioactively contaminated soil to reduce the contaminated soil volume. Many effective and safe remediation methods are available for restoring the sites of decommissioned nuclear facilities and realizing a sustainable environment.

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