Review Article

Review on Selection and Experiment Method of Commonly Studied Simulated Radionuclides in Researches of Nuclear Waste Solidification

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Although many types of simulated radionuclides have been widely used as a substitute for actual nuclear waste in the studies of nuclear waste solidification, the understanding of the applicability and validity of simulated radionuclides is still insufficient. In particular, the selection and use of simulated radionuclides, which can play a decisive role in the accuracy of the experimental results, still lack unified or integrated references. This paper provides a critical review on the selection, experimental methods, and applicability of the most commonly studied simulated radionuclides, followed by a careful discussion and recommendation of simulated radionuclides suitable for different solidified bodies. The main factors (e.g., temperature, pH, and atmosphere) affecting the choice of simulated radionuclides were analyzed in detail. This work helps to integrate the selection and use of simulated radionuclides, and it will be beneficial for improving the effectiveness of nuclide solidification research.

1. Introduction

Nuclear energy is a high-efficiency energy source with high energy density, low cost, and no air pollution emission [1]. However, the nuclear facilities of nuclear power, medicine, scientific research, industry, and agriculture can generate large amounts of nuclear waste during operation and decommissioning [2, 3]. Nuclear waste contains more than 30 radioactive elements, mainly including actinides (such as Pu and U), fission elements (such as Cs, Sr, I, Xe, and Ru), and other radioactive elements, which can emit radioactive alpha rays (α-rays), beta rays (β-rays), and gamma rays (γ-rays) during the decay of the nucleus, and X-rays by atomic shell electron transitions [4–6]. Nuclear waste can cause irreversible radioactive contamination to air, land, and water due to the high radioactivity and radiotoxicity [7]. When the human body is exposed to high doses of radiation, radioactive substances can cause irreversible damage to the human body, causing mutations in human cells and cancer [7, 8]. Based on two parameters, the radionuclide half-life and radioactivity, the International Atomic Energy Agency (IAEA) classified radioactive waste into five categories: very short-lived waste (VSLW), very low-level waste (VLLW), low-level waste (LLW), intermediate-level waste (ILW), and high-level waste (HLW) [7]. The main sources and types of radionuclides in nuclear waste are shown in Table 1. It can be seen that radionuclides and their isotopes are widely distributed in facilities, water, waste residues, and other wastes.

It is necessary to properly treat and dispose of nuclear waste for long-term safe and cost-effective management. Among the various nuclear waste management procedures, the immobilization treatment and geological disposal
methods are most widely used [7, 9]. Concrete and cement composites are widely used for LLW and ILW immobilization due to their good physical, chemical, and mechanical properties as well as low prices [10–12]. The common method for HLW immobilization is to form borosilicate glass or ceramic composites by vitrification or the melt process [8, 13]. Many studies [14–18] have been conducted to investigate the immobilization characteristics, mechanism, and effectiveness of various solidifying materials on nuclear waste. The main purposes of these studies are summarized as developing new solidifying materials, increasing the loading of radioactive waste, reducing the leaching of radionuclides, enhancing the mechanical strength of solidified bodies, and reducing the disposal difficulty and cost. Additionally, computer simulation methods [13, 19–23], such as quantum mechanical and empirical models of atomic bonding, density functional theory plus Hubbard U correction (DFT + U), energy minimization, molecular dynamics (MD), and Monte Carlo (MD) methods, have also been widely applied to study the radiation effects in various solidified bodies.

In most cases, actual nuclear wastes could not be used in these studies due to their radioactive, corrosive, and other harmful properties to the human body. In addition, it is impractical to study the radiation damage to the solidified bodies for hundreds of years or even thousands of years through actual experiments [2, 24]. Researchers [24–27] found that some nonradioactive isotopes of the radionuclides or some other nonradioactive elements had similar physical and chemical properties to the corresponding radionuclides. These nonradioactive isotopes or elements have been widely used as the simulated radionuclides to test or evaluate the solidification effect of solidifying materials on nuclear waste. Some simulated radionuclides commonly used in existing studies are shown in Table 2.

Table 1: Main sources and types of radionuclides [4, 6].

| Sources | Radionuclide | Form | Type | Waste form |
|---------|--------------|------|------|------------|
| Uranium mining and manufacturing | 226Ra, 238U, 234U, 235U | Solid | α, γ | Waste ore, resin, plastic, tailings, residue, waste equipment |
| | 234U, 235U, 236U, 238U, 232Th, 229Ra | Liquid | α, γ | Mine water, extraction water, purification waste liquid, wastewater |
| | 238U, 234U, 235U, 222Rn | Gaseous | α | Exhaust gas, dust, radioactive aerosol |
| | 137Cs, 144Ce, 239Pu, 129I, 90Sr, 133Xe, 135Xe, 239Np, 97Kr, 85Kr, 90Y, 99Tc, 131I, 103Ru, 106Ru, 99Tc | Solid and liquid | α, β, γ | Radioactive resin, mud, filter residue, waste equipment, waste tools, waste materials, cooling water, circulating water, ground drainage, exhaust gas, radioactive aerosol |
| Reactor operation, spent fuel | 60Co, 58Co, 16N | Solid | α, β, γ | Reactor vessel, waste core component, cladding material, graphite, waste equipment, concrete, circulating cooling water, decontamination wastewater |
| | 137Cs, 129I, 137Cs, 90Sr | Gaseous | α, β, γ | Exhaust gas, radioactive aerosol |
| Reactor operating facility | 60Co, 129I, 137Cs, 90Sr | Liquid | α, β, γ | Waste radioactive sources, radioisotopes, animal and plant pollutants, waste equipment, wastewater |
| Medicine, scientific research, industry, and agriculture utilization | 137Cs, 238U, 147Pm, 90Sr, 89Sr | Liquid | α, β, γ | Waste radioactive sources, radioisotopes, animal and plant pollutants, waste equipment, wastewater |
Radionuclides | Simulated nuclides and source compounds | Immobilization scheme
--- | --- | ---
$^{137}$Cs | Cs$^+$—CsNO$_3$, CsCl, CsOH, Cs$_2$CO$_3$ [10, 15, 28, 29] | Immobilization in cement, pollucite, glass, ceramic, polymer, geopolymer, inorganic mineral, etc.
$^{90}$Sr | Sr$^{2+}$—SrCl$_2$·6H$_2$O, Sr(OH)$_2$, SrCO$_3$, SrSO$_4$, SrF$_2$ | |
$^{60}$Co | Co$^{2+}$—Co(NO$_3$)$_2$·6H$_2$O, CoCl$_2$·6H$_2$O [29, 35, 36] | |
$^{239}$Pu | Pu$^{4+}$, Ce$^{4+}$—PuO$_2$, CeO$_2$ [24, 25, 37] | |
$^{129}$I | I$^-$, IO$_3^-$—KI, KIO$_3$, NaI, NH$_4$I, NH$_4$IO$_3$ [35, 38, 39] | |

However, despite common usage worldwide, there are no unified or integrated references for the selection and experimental method of the commonly studied simulated radionuclides.

This paper is oriented to review the selection, kinds, and experimental methods of simulated radionuclides commonly studied in the research of nuclear waste solidification. The advantages and disadvantages of the simulated radionuclides in different solidification conditions are discussed. The primary objective of the work is to (i) advance the understanding of various solidification approaches of nuclear waste and the role of simulated radionuclides and (ii) provide comprehensive references for the selection and use of simulated radionuclides in different solidification materials for nuclear waste solidification studies.

### 2. Simulants of $^{137}$Cs and Experimental Methods

Fission product $^{137}$Cs is one of the most important artificial radionuclides generated from nuclear reactors, nuclear weapons tests, radioactive wastes, or waste liquid [20]. It is a gamma emitter with radiotoxicity, high volatility, mobility, weapon tests, radioactive wastes, or waste liquid [20]. It is a gamma emitter with radiotoxicity, high volatility, mobility, and a relatively long half-life (close to 30 years) [47], which can be present in LLW, ILW, and HLW [48]. When $^{137}$Cs permeates into the food chain and is transferred to the human body, it can cause respiratory disease, acute and chronic injury, and even soft tissue tumors. Therefore, nuclear waste containing $^{137}$Cs must be pretreated and stabilized prior to disposal for the sake of preventing it from leaching into the environment [27].

$^{137}$Cs has been classified as an easy-to-measure nuclide, which can be used as the signal nuclide for nuclear power plant accidents [49]. It is reported that solid-phase or liquid-phase $^{137}$Cs exists in an oxide form such as Cs$_2$UO$_4$, Cs$_2$ZrO$_3$, or Cs$_2$MoO$_4$, and Cs$_2$SnO$_3$ in spent fuels [50–52], and the gaseous-phase Cs is considered to exist in Cs$_2$CO$_3$ (g), CsOH (g), and CsI (g) [53–55]. Thus, many studies were conducted to investigate the immobilization characteristics of solid-phase and liquid-phase $^{137}$Cs, and Cs$^+$ was the most used simulant of $^{137}$Cs due to the similarity in physical and chemical properties.

#### 2.1. Simulated $^{137}$Cs in the Immobilization Method by Using Cementitious Material

Cementitious materials have been widely used for radioactive waste solidification as long-term storage and disposal materials due to the good physical, chemical, and mechanical properties [11, 28, 56, 57]. Since the cement-based systems have presented the gradual dissolution and leaching of radionuclides, many studies have been conducted to improve this stabilization technique [58–61]. CsNO$_3$ or CsCl was often used as the simulant of $^{137}$Cs in those studies of cementitious material immobilization methods. The experimental methods of cementitious material solidification experiments for $^{137}$Cs are relatively simple and usually include the dissolution of simulated $^{137}$Cs, solidification, leaching tests, chemical tests, and mechanical performance tests.

Fan et al. investigated the effects of ferrihydrite nanoparticle incorporation in cementitious materials on radioactive waste immobilization [10]. In the experimental process, the cement-based solidified body for simulated $^{137}$Cs, $^{238}$U, and $^{90}$Sr was prepared first. The mass proportion of each simulated radionuclide in the solidified body was set as approximately 4%. First, CsNO$_3$ (0.72 g), UO$_2$(NO$_3$)$_2$·6H$_2$O (1.85 g), and Sr(NO$_3$)$_2$ (0.78 g) were dissolved in 20 mL of ultrapure water to prepare the radioactive waste solution. Then, 20 mL of simulated solution and 5 mL of suspension were added into 50 g of Type I ordinary Portland cement and mixed to reach a well-dispersed coherent state. The mixture was cast into a cylinder mold, demolded after 24 h, and then cured in at 20 ± 1°C and a relative humidity (RH) of 45 ± 5% for 28 days before the leaching test. In the end, the leaching test was conducted following the standard method of ANSI/ANS-16.1-2003R2008 (American National Standard Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Test Procedure). In another study [28], CsNO$_3$ was added into the used matrix (ordinary Portland cement blended with blast furnace slag), and the mass proportion of Cs was 3% in the total matrix.

In the study of the immobilization of the simulated radionuclide $^{135}$Cs$^+$ by fly ash-based geopolymer [15], CsNO$_3$ (analytical reagent grade) was used as the simulated radionuclide. Before being mixed with ordinary Portland cement and fly ash in proportion, CsNO$_3$ was first dissolved in deionized water to form a homogeneous solution. The mass proportion of Cs in the resulting geopolymer/cement mixture was 2%. The mixtures were made into 20 mm × 20 mm × 20 mm cubes and then cured in humid air with a relative humidity (RH) of 90 ± 1% and a temperature of 60 ± 0.5°C. In addition to microstructural and mineralogical analysis, leaching tests (long-term leaching, simulated environment leaching, and acid corrosion tests) and mechanical performance tests (compressive strength, freeze-thaw performance, and high-temperature performance tests) were also conducted to study the performance of the solidified body. Jang et al. used similar experimental
methods to study the physical barrier effect of the geopolymer waste form on the diffusivity of Cs and Sr [62], and the difference was that the nuclide simulants were CsCl and SrCl₂·6H₂O.

Cs⁺-compounds, which have the properties of water solubility, room temperature stability, and low toxicity, which can be selected as the simulants of ¹³⁷Cs in the experiments. According to former studies, the mass proportion of Cs⁺ in the cementitious mixture was no more than 5%, and ICP-MS or ICP-OES was usually used to quantitatively analyze Cs⁺ in the leaching test.

The advantage of CsNO₃ and CsCl is that their aqueous solutions are close to neutral, and thus, they have less of an effect on cement solidification. One challenge associated with CsNO₃ and CsCl is matching the amount dissolved in water with the water–cement ratio during the solidification experiments.

2.2. Simulated ¹³⁷Cs in the Immobilization Method by Using Pollucite. Pollucite belongs to the group of analcime zeolites, and the specific structure of pollucite is beneficial to hold Cs ions tightly and avoid leaching in aqueous media [63, 64]. The thermal stability of pollucite has been confirmed to be excellent relative to several cesium aluminosilicate minerals [65]. Therefore, pollucite (CsAlSi₂O₆) may be feasible as one of the final phases to dispose of ¹³⁷Cs-containing waste for extended periods of time [66–68]. Pollucite can be synthesized by the hydrothermal route (Figure S1) or recrystallized by heat treatment [69–71].

Chen et al. investigated the method of the hydrothermal conversion of Cs-polluted soil into pollucite for ¹³⁷Cs immobilization [18]. In the experiment, alumina was mixed with ground soil to form certain molar ratios of Al/Si, and then, 5 mass% CsOH solutions were added into the mixture at Cs/Al/Si ratios of 1/1/5, 1/1.5/5, 1/2/5, and 1/2.5/5, respectively, in order to synthesize pollucite. The specimens were pressed in a stainless rectangular-shaped mold and then cured for various setting times (0–24 h) in a Teflon-lined stainless steel autoclave at setting temperatures (100–240°C) for pollucite preparation. The pollucite (CsAlSi₂O₆·xH₂O) samples showed low flexural strength (5 MPa). With the addition of Ca(OH)₂, a strong solidified body was synthesized, which showed the highest flexural strength of 30 MPa. EXAFS measurements were performed to investigate the coordination environments of the synthesized pollucite and further confirm the coordination state of Cs in the synthesized pollucite structure. According to the EXAFS test, the polluted Cs was immobilized into the structure of the synthesized pollucite. The leaching test of Cs in pollucite samples was conducted according to ASTM C 1285-02 (American Society for Testing Material, 2008) [72], rather than ANSI/ANS-16.1-2003R2008. The ASTM C 1285-02 method is suitable for glass waste forms or multilayer glass ceramics [72]. Pollucite can be converted into ceramic by calcination and is also suitable for this leaching test method. This hydrothermal technology showed potential to directly immobilize the ¹³⁷Cs-polluted soil.

Montagna et al. also studied the storage of ¹³⁷Cs by borosilicate and aluminosilicate pollucite nanocrystals [48]. CsOH, silica sources (precipitation silica Zeosil 175MP Rhône-Poulenc or fumed silica Aerosil 200), H₃BO₃, NaOH, and aluminum (AlOOH) hydroxides were mixed at certain molar ratios (0.22 ≤ (B + Al)/Si ≤ 0.43, 0 ≤ B/(B + Al) ≤ 1, and 0.10 ≤ Cs/(Na + Cs) ≤ 1). The mixtures were sealed in a stainless steel autoclave and then heated for 2–30 days at 115–195°C to prepare pollucite crystalline materials.

The process of Cs-pollucite preparation is relatively simple. Similar methods were also applied in other researches [67, 70, 73, 74], and the pollucite was verified to incorporate Cs into the crystalline structure, which presented an excellent immobilization effect on Cs. The advantage of CsOH is that it does not introduce any other ions except for O and H into the synthesis reaction of pollucite, and the reaction products are only pollucite and H₂O, which is conducive to the purity of the prepared pollucite. However, CsOH is highly corrosive and should be placed in a sealed container that is stored in a cool, dry place.

2.3. Simulated ¹³⁷Cs in the Immobilization Method by Using Glass Waste Forms. Glass waste forms have the potential to provide durable, wide-ranging, and good radiation resistance immobilization of HLW [26, 75, 76]. Sohn et al. studied the stability of Cs and Sr in Na-aluminoborosilicate glasses [77]. In this study, three glass compositions were prepared based on the following systems: Na₂O·Al₂O₃·B₂O₃·SiO₂·Cs₂O (system NABS-Cs), Na₂O·Al₂O₃·B₃O₃·SiO₂·SrO (system NABS-Sr), and Na₂O·Al₂O₃·B₂O₃·SiO₂·SrO·Cs₂O·SrO (system NABS-SrCs). The preparation process of glass samples is described as follows: (i) simulated nuclides (Cs₂CO₃ and SrCO₃) and reagent-grade Na₂CO₃ were preheated and decarbonated at 850°C for over 30 minutes; (ii) reagent-grade B₂O₃, Al₂O₃, and SiO₂ were dried at 1200°C for over 20 minutes; (iii) these reagents were weighed and mixed according to the set ratios and then melted in a platinum crucible at 1200°C for 1 h; (iv) the melt was quenched, and the obtained glass was crushed and ground; and (v) the melting and grinding process was conducted twice. This experimental method enabled the preparation of glass samples with a fairly homogeneous chemical composition, and most simulated nuclides were immobilized in crystalline materials during the devitrification of the glasses. The prepared glasses were analyzed by a JEOL 733 electron microprobe to check the chemical change by melting. The analyzed mass proportions of Cs₂O and SrO were 4.54%–13.71% and 4.97%–10.44%, respectively, in the aluminoborosilicate glasses. The results showed that the Cs₂O content in the samples decreased markedly (2%–3%) because Cs₂CO₃ was partially decomposed and volatilized during melting. Compared with Cs₂O, the SrO content was almost unchanged.

To investigate the influence of the hydrothermal reaction between glass and water on Cs and Sr immobilization, this study conducted hydrothermal treatment of the prepared glass samples. The powdered glass samples were sealed in a gold capsule with deionized water and then heated in an autoclave at 200°C with a vapor pressure of 1.54 MPa for 60 days. The heating temperature was
determined according to the maximum temperature (150–200°C) in the deep geological repositories in the first few hundred years after disposal [78]. The produced crystalline phases were identified as Analcime-type zeolite by the XRD method. Cs and Sr were found to be fixed in the analcime-type zeolite (Figure S2). The experimental methods in this study have great reference value for the preparation method of $^{137}\text{Cs}^+$ (or $^{90}\text{Sr}^+$) containing glasses as well as the hydrothermal treatment of the glasses for the hydrothermal reaction study. Similar methods were also used in related studies [79–82].

In mixed alkali glasses (e.g., Na-K or Na-Cs silicate glasses), physical properties such as the electrical conductivity, molar volume, glass transition temperature, and thermal expansion coefficient can be nonlinear due to the mixed alkali effect (MAE) [83, 84]. The MAE has attracted much attention because it is very important for developing unique glass materials with a controllable electrical conductivity or thermal expansion coefficient, respectively [19, 85]. Kaneko et al. [19] prepared borosilicate (Si:B = 1:1) and Si-rich borosilicate (Si:B = 2:1) glasses containing alkali cations Cs$^+$ and Na$^+$ to study the MAE on nuclides solidification. In the preparation process of the glass samples, chemically pure Cs$_2$CO$_3$ together with SiO$_2$, B(OH)$_3$, and Na$_2$CO$_3$ were used to prepare the glass samples by the melting method (temperatures of 1350°C, 1100°C, and 1350°C for silicate, borate, and borosilicate glasses, resp.). The Cs$^+$/ (Cs$^+$+Na$^+$) ratios were set at 0.0, 0.2, 0.5, 0.8, and 1.0. XRD was used to identify the crystal structures of the synthesized samples. A strong magnetic field MAS NMR was used to examine the structure of mixed alkali borosilicate glasses for the sake of correlating the MAE with the alkali environment. An AVANCE III spectrometer using a commercial probe (4 mm) was used to acquire the solid-state $^{133}\text{Cs}$ and $^{23}$Na NMR spectra of all samples. According to the NMR analysis, the chemical shifts of alkali ions are affected by both the coordination number and the kinds of nearby atoms (Si or B). The coordination characteristics of Cs$^+$ in the crystal of borosilicate glasses could help to study the immobilization mechanism of $^{137}\text{Cs}$ in the actual glass solidified body (Figure S3). This experimental method was effective for studying the MAE in Cs-containing glasses, and similar methods were also used in other studies [86, 87]. It can be seen that the high-temperature calcination process (1100–1350°C) is an essential process for glass preparation. In the typical preparation process of borosilicate glass, Cs$_2$CO$_3$ and Na$_2$CO$_3$ could offer Cs$_2$O and Na$_2$O for the SiO$_2$-B$_2$O$_3$-R$_2$O composition system (R$_2$O refers to Na$_2$O, K$_2$O, or Cs$_2$O) during the high-temperature solid-phase reaction. The NMR and XRD analysis results in the above studies confirmed that Cs$^+$ could enter the crystalline structure of the prepared glasses and successfully simulated the immobilization process of $^{137}\text{Cs}$ in the glass solidified bodies. The high temperature may inevitably cause some volatilization loss of Cs$_2$O in the glass [77], and this can lead to numerical deviations in the quantitative analysis of Cs$^+$. Thus, the residual Cs$^+$ content in the prepared glass samples needs to be determined in some quantitative studies, such as the leaching test of the glass solidified body.

Different from the synthesis of pollucite, CsOH could not be used as the raw material of glass because of its very strong alkali nature. It may react violently with the metal containers and corrode the glasses at high temperature, which will make the experiment unsafe.

2.4. Simulated $^{137}\text{Cs}$ in the Immobilization Method by Using Ceramic Waste Forms. Ceramic matrices exhibited more remarkable radiation resistance and chemical durability than glasses, and they were considered as the ideal hosts for immobilizing HLW [8, 44, 45]. In Yang et al.’s study [27], the (Cs, Ba)-hollandite ceramics were prepared to study the chemical evolution in the hollandite matrix due to radioactive decay ($^{137}\text{Cs} \rightarrow ^{137}\text{Ba}$), and nonradioactive $^{133}\text{Cs}$ and $^{133}\text{Ba}$ were chosen as the simulants of parent $^{137}\text{Cs}$ and daughter $^{137}\text{Ba}$, respectively. The $[\text{Cs}_x\text{Ba}_y]_2[\text{Ti}_4\text{O}_{12}]_2\text{H}_2\text{O}$ (0.3 < x, y < 0.75) ceramics were designed for $^{137}\text{Cs}$ and $^{137}\text{Ba}$ immobilization, and they were synthesized by a solid-state reaction method. The raw materials were Cs$_2$CO$_3$, BaCO$_3$, TiO$_2$, and TiO$_2$, among which TiO$_2$ was introduced to conserve the charge neutrality when Cs$^+$ ions were replaced by Ba$^{2+}$ ions. The materials were preheated and weighed based on the composition of $[\text{Cs}_x\text{Ba}_y]_2[\text{Ti}_4\text{O}_{12}]_2\text{H}_2\text{O}$ (0.3 < x, y < 0.75) ceramics. Then, they were homogenized in an agate mortar in ethyl alcohol by grinding and mixing for 3 h. The mixtures were pressed into discs (12 mm in diameter, 2 mm in thickness) at a pressure of 12 MPa by using a hydraulic press. Then, the as-prepared discs were sintered in 5% H$_2$/Ar at 1250°C for 4 h to prepare the ceramic samples. Some other studies [14, 23, 88, 89] also used similar reagent-grade oxide, carbonate powders, and melt methods to prepare hollandite waste forms.

The phase composition and evolution of the ceramics were analyzed by XRD, and the microstructure evolution was detected by a Renishaw inVia Raman spectrometer, field scanning electron microscopy (FSEM), and transmission electron microscopy (TEM) coupled with energy-dispersive X-ray spectroscopy (EDX). The EDX mapping images of the prepared [CS$_{0.55}$Ba$_{0.5}$][Ti$_2$O$_3$]O$_{16}$ ceramic sample are shown in Figure S4 (e1, e2, e3, and e4), and all of the elements Cs, Ba, Ti, and O were found to be homogenously distributed on the sample surface. Furthermore, the EDX spectra in Figure S4 (f1 and f2) indicated that the experimental compositions of the ceramic samples were very close to the targeted compositions, and more than 80% of Cs was retained in the ceramic sample. The results indicated that the experimental method in this study could prepare Cs, Ba-containing ceramic samples with uniform element distribution and low nuclide loss for nuclide solidification research.

This study used the following equation to estimate the structural stability: $t_H = ((t_f + r_O)/(r_B + r_O))^{1/2}/\sqrt{3}/2$, where $t_H$ is the tolerance factor, $r_A$ is the average ionic radii of cations in the A site, $r_B$ is the average ionic radii of cations in the B site, and $r_O$ is the oxygen ion radius (1.4 Å). The calculation results showed that the value of $t_H$ was close to 1 (1.033–1.097), which
suggested that the crystalline structure of the (Cs, Ba)-
hollandite ceramics was stable. A diagram of the crystal
structure was used to depict the evolution of simulated 137 Cs
decay to simulated 137 Ba (Figure S5), which showed that the
nuclides were chemically immobilized in the ceramic crystal.
Overall, this study combined a Cs, Ba-containing ceramic
preparation experiment, mathematical calculations, and
crystal model construction, which provided comprehensive
experimental methods for analyzing the immobilization and
decay characteristics of simulated 137 Cs in the ceramic system.

In another study [20], which investigated the effects of Cs
substitution on the crystal structure of phase-pure hollandite
of the system, Ba1.15−xCs2xCr2.3Ti5.7O16 (0 ≤ x ≤ 1.15), CsNO3,
Ba(O2C2H3)2, Cr(NO3)3·9H2O, and Ti(OC2H5)4 were used as
starting materials for the preparation of the hollandite samples.Unlike Yang et al.'s study, the starting materials
were dissolved individually in a suitable solvent and ho-
mogenized by mixing the solutions. The resultant solution
was concentrated in a rotary evaporator at 90°C and was then
calcined at 400°C for 4 h to remove all of the volatile or-
ganics. The calcined powder was pressed into discs (20 mm
diameter, 3 mm thickness) at a pressure of 35 MPa for
subsequent sintering at 1050°C/4 h. The material mixing
method in this study can make the components more ho-
mogeneous, but the pretreatment process for the mixture
before calcination is more complicated.

In addition to XRD analysis, time-of-flight (TOF)
neutron powder diffraction (NPD) experiments were also
performed for the structural analysis of phase-pure samples
in this study. Furthermore, Rietveld refinement of XRD and
NPD data was carried out to extract the structural features.
Powder XRD data was refined by using the TOPAS 4.2
(Bruker AXS, Karlsruhe, Germany) software package in the
GUI mode, starting with the tetragonal (I4/m) and mono-
clinic (I2/m) structural models. Figure S6 shows a standard
mixture was heated in the temperature range of 750–1200°C
solutions and metakaolin were mixed to form a homoge-

eous paste, which was subsequently cast in sealed con-
tainers cured for 3 months at either 20 ± 2°C or 80 ± 2°C. The obtained molar ratio of Sr/Al was 0.00025, which
was representative of the waste forms produced by the cemen-
tation of radioactive ion exchange resins [31, 93]. Sr(OH)2 is
easily soluble in water and can be completely ionized, and
that is why it was chosen as the experimental material.

In addition to Sr(NO3)2 [94, 95], SrCO3 [33], and
Sr(OH)2, SrSO4 [30] and SrF2 [34] were used to prepare glass
or ceramic waste forms with other materials in some studies
due to their high melting points (>1400°C). The application
conditions and advantages of the simulants of 90 Sr in the
present studies are shown in Table 5.

4. Simulants of Some Other Nuclides and
Experimental Methods

Except for 137 Cs and 90 Sr, which have been widely studied,
some other radionuclides such as 239 Pu, 60 Co, and 129 I are
also common nuclides in nuclear waste [96, 97]. These ra-
dionuclides are usually distributed in HMW and MMW,
which have strong radiation hazards [35, 98]. It is important
to pretreat and stabilize these elements prior to disposal.
Therefore, many studies have been conducted to investigate
the solidification/stabilization method and mechanism.

4.1. Simulants of 239 Pu.

The safe disposal of radioactive actinides (239 Pu, 230 Th, 232 U, etc.) generated by the nuclear fuel cycle has been a major topic in the nuclear industry
due to their long-term radiotoxic contribution to HLW
[26, 46]. Ceramics are considered to be suitable host

3. Simulants of 90 Sr and Experimental Methods

90 Sr is a very common beta-emitter in nuclear installations
[30], which exists in wastewater, ion-selective inorganic
resins, waste soils, and so on [91]. It can easily migrate into
ground water due to its water solubility, and thus, it should
be stabilized to prevent it from polluting the environment.
Sr2+ is the main simulant of 90 Sr in present studies of 90 Sr
immobilization [32, 33, 62]. In some of the studies men-
tioned above [10, 62, 77], 90 Sr was studied simultaneously
with 137 Cs in the same solidified bodies in which Sr(NO3)2,
SrCO3, and SrCl2·6H2O were used as the simulants of 90 Sr.

In the study of the immobilization of Sr by crystalline
zirconium phosphate [32], Sr(NO3)2 was added to
HZr2(PO4)3 (molar ratios: 0.1–0.5) and mixed, and then
the mixture was heated in the temperature range of 750–1200°C
for the preparation of the SrZr2(PO4)3 solidified body. In
addition to conventional composition and structure analysis,
the leaching characteristics of Sr in various solvents
(0.1 mol/L HCl, deionized water and sea water) in an au-
toclave at 160°C, were also investigated. The leaching test
results indicated that the crystalline proton type zirconium
phosphate had excellent Sr2+ leaching resistance, which was
thermally and chemically resistant in different leaching
environments. In the heating process, Sr(NO3)2 could de-
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materials for the immobilization of minor actinides [99–101]. Plutonium (Pu) is a representative actinide existing in HMW, and there has been extensive interest in using materials to immobilize it [46, 96, 102]. Kulkarni et al. prepared Pu-pyrochlore, \((\text{La}_{1-x}\text{Pu}_x)\text{Zr}_2\text{O}_7\) \((x \approx 0.15)\), as the ceramic wasteform for \(^{239}\text{Pu}\) immobilization [25]. \(\text{PuO}_2\), \(\text{ZrO}_2\), graphite, and \(\text{La}_2\text{O}_3\) were mixed in appropriate ratios and heated up to 1400°C in a helium atmosphere to prepare the Pu-pyrochlore. x\(\text{Qhe}\) TG and DTA analysis results of the \(2\text{PuO}_2 + 2\text{ZrO}_2 + 2\text{C}\) mixture are shown in Figure S7. x\(\text{Qhe}\) high-temperature chemical reaction was slow, extending over a temperature range of 1000–1400°C associated with a broad endothermic peak. x\(\text{Qhe}\) XRD analysis of the product indicated that it was \(\text{Pu}_2\text{Zr}_2\text{O}_7\), the XRD pattern of which was similar to that of \(\text{La}_2\text{Zr}_2\text{O}_7\). According to the XRD, TG, and DTA results, the chemical reaction could be given as follows: \(2\text{PuO}_2 + 2\text{ZrO}_2 + 2\text{C} \rightarrow \text{Pu}_2\text{Zr}_2\text{O}_7 + 2\text{CO}\). It could be concluded that the immobilization effect of Pu-pyrochlore on Pu is mainly a chemical immobilization based on the Pu ion entering the lattice of pyrochlore. Burakov and Anderson also used \(\text{PuO}_2\) to prepare Pu-pyrochlore for the durability study of actinide ceramic waste forms [37]. x\(\text{Qhe}\) prepared samples showed highly physicochemical resistance to ion-saturated solutions simulating underground water after alteration experiments at 90°C for 3 months. \(\text{PuO}_2\) can be used as the simulant of \(^{239}\text{Pu}\) in the studies of nuclide solidification. However, the challenge is that \(\text{PuO}_2\) is highly toxic and has strong radioactivity, and thus, it is not proper to be used in actual experiments [103]. Many studies used \(\text{Ce}^{4+}\) as a substitute for \(\text{Pu}^{4+}\) because they have the same chemical valence state and similar ionic radii and chemical properties, and \(\text{CeO}_2\) has much lower toxicity and radioactivity than \(\text{PuO}_2\) [24, 46, 104–106].

| Table 3: Unit cell dimensions and fit parameters from the Rietveld refinement of \(\text{Ba}_4\text{C}_{1.15−x}\text{Cs}_x\text{Cr}_{2.3}\text{Ti}_{15−x}\text{O}_{16}\). |
|----------------|-------------|-------------|-------------|
| Composition   | x = 0      | x = 0.15    | x = 0.25    |
| Space group   | \(I4/m\)   | \(I2/m\)    | \(I2/m\)    |
| \(a (\text{	extdegree})\) | 10.0561 (2) | 10.0244 (6) | 10.192 (1)  |
| \(b (\text{	extdegree})\) | —         | 2.9333 (2)  | 2.911 (0)   |
| \(c (\text{	extdegree})\) | 2.95210 (7) | 10.0075 (6) | 10.159 (1)  |
| \(\beta\)     | 90.058 (2) | 90.126 (3)  | 90.126 (3)  |
| Volume, \(V (\text{\textdegree})\) | 298.53 (2) | 298.95 (3)  | 303.34 (7)  |
| \(d, \text{\textdegree}\) | 5.08       | 5.09        | 5.18        |
| Refinement residual | \(R_{wp}\) (%) | 3.72        | 3.68        |

“\(d\)” is the mean diagonal length of the tunnel cross section, and “\(R_{wp}\)” is the weighted Rietveld parameter [20].

| Table 4: The application conditions and advantages of the simulated \(^{137}\text{Cs}\). |
| Simulants           | Solidified body     | Experimental method | Advantages                                                                 |
|---------------------|---------------------|---------------------|-----------------------------------------------------------------------------|
| \(\text{CsNO}_3\) or \(\text{CsCl}\) | Cementitious materials | Dissolved in deionized water and directly mixed in the solidified body for immobilization (<100°C) | Stable at low temperature, soluble in water, easy to obtain |
| \(\text{CsOH}\)     | Pollucite            | Hydrothermal reaction (100–200°C) | OH− in \(\text{CsOH}\) can be converted to \(\text{H}_2\text{O}\) by reaction; no other elements were introduced into pollucite except for Cs |
| \(\text{Cs}_2\text{CO}_3\) or \(\text{CsNO}_3\) | Glass or ceramic     | Melting method (1000–1350°C) | Decompose at high temperature, releasing \(\text{Cs}_2\text{O}\) for the solid-phase reaction; \(\text{CsOH}\) is not used because it is strongly alkaline and will react violently with the metal container |

| Table 5: The application conditions and advantages of the simulated \(^{90}\text{Sr}\). |
| Simulants           | Solidified body     | Experimental method | Advantages                                                                 |
|---------------------|---------------------|---------------------|-----------------------------------------------------------------------------|
| \(\text{SrCl}_2\cdot 6\text{H}_2\text{O}, \text{Sr(NO}_3)_2\) | Cementitious materials | Directly mixed in the solidified body for immobilization (<100°C) | Soluble in water; the aqueous solution is neutral and does not affect hydration |
| \(\text{Sr(OH)}_2\)  | Geopolymer gel      | Directly mixed in the solidified body for immobilization (<100°C) | Easily soluble in water, completely ionized, no other elements were introduced except for Sr |
| \(\text{Sr}_2\text{O}_3\) or \(\text{SrF}_2\) | Glass or ceramic     | Melting method (1000–1350°C) | High melting points (>1400°C), participates in solid-phase reaction at high temperature to provide \(\text{SrO}\) for the product (\(\text{SrF}_2\) converts to \(\text{SrO}\) at >1000°C in air) |
| \(\text{Sr(NO}_3)_2\) | Phosphate or geopolymer | Melting method (750–1200°C) | Decomposes at high temperature (melting point is 570°C), releasing \(\text{SrO}\) for the solid-phase reaction |

The presented samples showed highly physicochemical resistance to ion-saturated solutions simulating underground water after alteration experiments at 90°C for 3 months.
1673 K for 32h to prepare (La1−xCe0.5)Zr2O7. Then, the prepared powders were reduced, treated, and sealed under vacuum containing residual He in quartz vials. The purpose of the reduction treatment was to reduce Ce(IV) to Ce(III). This is because Ce(IV) can cause a structural change from the ordered pyrochlore to a disordered defect fluorite due to the smaller ionic size (0.097 nm) compared to that of Ce(III) (0.114 nm). Ce(III) is more stable than Ce(IV) in the vacuum environment, and a single-phase stable Ce-pyrochlore is beneficial to obtaining precise experimental results. The microstructural evolution of the Ce-doped La2Zr2O7 and Ce0.5Zr2O5 upon ion beam irradiation was characterized by in situ transmission electron microscopy (TEM), ex situ high-resolution TEM, and electron energy-loss spectrometry (EELS) techniques. According to the results, the Ce4+ fraction was estimated to be ~30%, suggesting that Ce3+ mainly dominated in the pyrochlore structure upon ion irradiation.

4.2. Simulants of 60Co. 60Co is an isotope of cobalt (Co), which is mainly generated during the operation of nuclear reactors. It has high penetrating gamma radiation (Eγ: 1.17 MeV and 1.33 MeV) and a half-life of 5.26 years [36]. It mainly exists in the coolant, which is a source of serious radiation exposure to the radiation workers during normal operation [107]. Once it has entered the environment, it may be absorbed by soil or react with particles and cause health effects [108].

Some researchers studied the immobilization of 60Co by using hydroxyapatite for 60Co-containing radioactive waste management. Ul Hassan et al. [16] studied the immobilization of Co-containing radioactive corrosion products by the cold sintering of calcium hydroxyapatite (HAp), and Co2+ was used to simulate 60Co. According to the previous study [108], the simulated radioactive solutions were prepared by dissolving Co(NO3)2·6H2O salt in distilled water and adjusting the initial pH values of the solutions to 5.0 ± 0.1. In the experiment, Co2+ was absorbed by HAp, which was prepared by mixing (NH4)2HPO4 and Ca(NO3)2·4H2O solutions. The absorbent (Co-HAp) slurries were then dried overnight at 110°C in a vacuum oven. The dried Co-HAp powder was sintered by applying the optimized sintering conditions (200°C, 100 min, and 500 MPa). The sketch of the experimental arrangement used for the cold sintering of Co-HAp is shown in Figure S8.

The measured specific surface area of the prepared HAp was 112.35 m²/g, which enabled a much higher adsorption capacity of HAp than those sorbents in some typical Co2+ adsorption studies [97, 108, 109]. The results showed that 0.4 g of the prepared HAp could absorb 93.8 ± 0.2% of 1 mM Co2+ in 100 ml of aqueous solution. After sintering at 200°C, the Co-HAp was tested by SEM-EDS for the microstructural characterization and elemental distribution analysis. The EDS-elemental area mapping showed that the absorbed Co2+ was homogeneously distributed in the sintered matrix. The normalized leaching rate (NLR) of the absorbed Co2+ in Co-HAp was then measured by a standard product consistency test (PCT). The calculated NLRi values of Co2+ were 2.5 × 10−3 g/m²/day, which is better than the release limit of 0.3 g/m²/day set by the Department of Energy (DOE) of USA for types of phosphate glass [110].

In conventional sintering methods [111, 112], HAp was sintered at high temperatures ranging from 900 to 1300°C in air. However, the high temperature can cause the decomposition or dihydroxylation of the HAp-based radioactive waste matrices [113] or the volatilization of radionuclides such as 129I [114]. The cold sintering technique of HAp for the immobilization of radioactive waste is more efficient, economical, and environmentally friendly.

Venkatesan et al. used a similar simulant and experimental method [36] as above. The advantage of using Co(NO3)2·6H2O as the source of Co2+ is that it will not affect the adsorption of Co2+ by HAp because NO3− does not further react with HAp to destroy its structure.

4.3. Simulated Radioiodine and Other Nuclear Waste. Radioactive iodine isotopes, principally 129I and 131I, are radiation hazards emanating from the waste stream of spent fuel reprocessing facilities, which have high mobility and long half-lives (exceeding 1 million years) [115]. 129I is consistently found to be one of the largest dose contributors in geologic repository performance assessments [98]. The capture and immobilization of 129I are necessary in the reprocessing facility for safe disposal on a geological time scale.

Riley et al. [98] have reviewed the immobilization methods and materials of radioiodine, and a wide range of sorbents for the capture and waste forms for the immobilization of radioiodine were summarized and assessed. As described in this review, various iodine species such as NaI, NaIO3, HgI2, and I2(g) were used as the simulants of 129I in the current literature. Depending on the solidification approach and what form of iodine is present in the waste forms, the iodine species used as simulants of radioiodine are different.

In addition to the iodine species mentioned above, NH4IO3 was used as the simulant of 129I in Hassan et al.’s study [35]. The cold sintering of iodine-hosted HAp for the immobilization of volatile radioiodine (I) was investigated. During the experiment, an anionic solution of [NH4IO3 + (NH4)2HPO4] was mixed with a cationic solution of Ca(NO3)2·4H2O dropwise at a flow rate of 3.3 ml, which formed a Ca:PH molar ratio of 10:6:2. Then, IO-HAp was synthesized at 70°C and dried for 12 h at 110°C. The cold sintering of the IO-HAp powder was carried out by applying the temperature of 200°C, a uniaxial pressure of 500 MPa, and a holding time of 10 min. According to the XRD, TGA/DSC, XRF, ICP-MS characterization, and PCT test of the IO-HAp powder samples, IO-HAp showed several advantages for the immobilization of 129I and other volatile radionuclides for long-term geological disposal, such as simple, fast, energy-saving, and efficient solidification. The advantage of NH4IO3 is that NH4+ could be released as NH3 during the synthesis reaction of IO-HAp, which will not affect the Ca:P: I molar ratio and the crystal structure of IO-HAp. However, NH3 is harmful to the experimenters, and that is why the
synthesis of IO-HAp was carried out in a fume hood under continuous stirring of 200 RPM [35].

Radioactive resins are problematic wastes that are produced during the ion exchange process for primary coolant purification and liquid radioactive waste treatment, which contain various radionuclides (e.g., $^{137}$Cs, $^{60}$Co, and $^{85}$Sr) [11]. Sun et al. [116] investigated the solidification of borate radioactive resins using sulfoaluminate cement blended with zeolite. In this study, the simulated borate radioactive resins were prepared by the following steps: (i) Amberlite INR 78 resin and INR 77 resin were saturated by $\text{H}_2\text{Bo}_3$ and $\text{NaOH}$, respectively, and then mixed in a mass ratio of 1:3 and (ii) $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and $\text{CsCl}$ were added in the resins as the simulated radionuclides, and the concentrations of $\text{Co}$ and $\text{Cs}$ in the blended resins were 3.963 g/L and 4.093 g/L, respectively. This preparation method for simulated radioactive resins is simple and easy to operate.

In Li et al.'s study [29], a new resin mixture of styrene base cation and anion exchange resins with a mixing ratio of 1:1 (by mass) was contacted with a known concentration of saturated solution containing $\text{CsCl}$ and $\text{CoCl}_2$ for a prolonged time (1 week). The solution contacting the resins was shaken several times every day. By this static method, the saturation of simulated resins was performed. Then, the resins were dehydrated until the moisture content decreased to 45–50% before cementation. The saturation process is a key step in the preparation of the simulated radioactive resins.

5. Summary and Conclusions

Simulated radionuclides are used to directly simulate the solidification and migration characteristics of the actual radionuclides in various solidified bodies. The aim of this work is to review the selection and experimental methods for the simulants of the most commonly studied radionuclides (e.g., $^{137}$Cs, $^{90}$Sr, $^{60}$Co, $^{239}$Pu, and $^{129}$I) in the research of nuclear waste solidification and provide the selection basis of commonly used simulated radionuclides in various application conditions.

It is found that the nonradioactive isotopes of radionuclides are preferred as the simulants of the corresponding radionuclides because of their similar ionic radii, chemical valence state, and some other physicochemical properties. It is suggested that the simulated radionuclides are suitable for the corresponding solidified bodies and immobilization methods. In addition, the compounds containing simulated radionuclides should be nontoxic or have low toxicity and be nonradioactive or have low radioactivity to reduce the health risk to the experimenters. For $^{137}$Cs solidification studies, $\text{Cs}^+$ (cesium salt) is mostly used as the simulated nuclide. $\text{CsNO}_3$, $\text{CsCl}$, $\text{CsOH}$, $\text{Cs}_2\text{CO}_3$, and $\text{CsNO}_3$ are the commonly used $\text{Cs}^+$-containing compounds. For $^{90}$Sr solidification studies, $\text{Sr}^{2+}$ (strontium salt) is mostly used as the simulated nuclide. $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$, $\text{Sr(NO}_3)_2$, $\text{SrCO}_3$, $\text{SrSO}_4$, or $\text{SrF}_2$ is chosen to be used as the source of $\text{Sr}^{2+}$. For $^{239}$Pu solidification studies, $\text{PuO}_2$ and $\text{CeO}_2$ are mostly used as the simulants in the ceramic solidification methods occurring at high temperature. $\text{Co(NO}_3)_2 \cdot 6\text{H}_2\text{O}$ is commonly used as the simulant of $^{60}$Co, and various iodine species such as $\text{NH}_4\text{IO}_3$, $\text{NaI}$, $\text{NaIO}_3$, $\text{HgI}_2$, and $\text{I}_2(\text{g})$ are used as the simulants of $^{129}$I. For radioactive resin solidification studies, simulated radionuclides such as $\text{CsCl}$ and $\text{CoCl}_2$ are added in resins for the preparation of simulated radioactive resins. Multiple simulated nuclides can be simultaneously used in the same nuclear waste solidification experiment.

The preparation conditions of the solidified body for radionuclides, such as the temperature, phase, atmosphere, reaction principle, etc., dominate the selection of the simulants. For different solidification methods and materials, some recommendations for the selection of simulated radionuclides are summarized as follows:

(i) For cementitious material geopolymer solidification methods and cold sintering methods, the compounds containing simulated radionuclides should be soluble in water and stable at low temperature (<200°C). Moreover, the pH value of the aqueous solution of the simulants is recommended to be close to that of the solidification environment, which will have less influence on the solidification process.

(ii) For hydrothermal methods and other cold sintering methods with a reaction temperature at ∼200°C, the simulated radionuclides should not introduce other elements or ions except for those mainly involved in the reactions. In this case, the reaction products containing simulated radionuclides are usually phase-pure for the sake of obtaining precise research results.

(iii) For glass, ceramic, or some other solidified bodies, which are prepared by the melting methods, the simulants of radionuclides should have physicochemical stability at room temperature and can decompose at high temperature (approximately 900°C–1400°C) to release the oxides of simulated radionuclides for the solid-phase reactions.

(iv) It is recommended that the simulated radionuclides are homogenized in the raw materials to increase the uniformity of the prepared solidified bodies, and this can improve the representativeness and validity of the test results.

This work will be beneficial in extending the applications of simulated radionuclides in new solidification materials and promoting the development of nuclear waste solidification techniques. However, there are still some other radionuclides that have not been reported in the research of nuclear waste solidification. Further studies will be conducted on the solidification methods and simulants of more radionuclides.

Data Availability

No data were used to support this study.

Conflicts of Interest

The authors declare that they have no conflicts of interest regarding the publication of this paper.
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Supplementary Materials

Figure S1: the hydrothermal route for Cs-pollutate preparation. Figure S2: distribution of cations between the starting glasses and formed zeolites. Figure S3: 133Cs MAS NMR spectra of 4βCs2O-4(1-β)Na2O-8SiO2-2B2O3 glasses. Figure S4: element mapping and EDX spectra of [Cs0.55Ba0.5]2-8SiO2-2B2O3 glasses. Figure S5: the substitution of Ba2+ for Cs2+ in the ceramic system. Figure S6: hollandite unit cell with I2/m symmetry. M1 and M2 are two distinct crystallography sites for octahedral cations (Cr and Ti). Figure S7: TG and DTA curves of 2PbO + 2ZrO2 + C in helium. Figure S8: experimental arrangement used for the cold sintering of Co-HAp. (Supplementary Materials)

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