Research Paper https://doi.org/10.7733/jnfcwt.2023.040

Preliminary Selection of Safety-Relevant Radionuclides for Long-Term Safety Assessment of Deep Geological Disposal of Spent Nuclear Fuel in South Korea

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(Received September 19, 2023 / Revised October 4, 2023 / Approved October 13, 2023)

With South Korea increasingly focusing on nuclear energy, the management of spent nuclear fuel has attracted considerable attention in South Korea. This study established a novel procedure for selecting safety-relevant radionuclides for long-term safety assessments of a deep geological repository in South Korea. Statistical evaluations were performed to identify the design basis reference spent nuclear fuels and evaluate the source term for up to one million years. Safety-relevant radionuclides were determined based on the half-life criteria, the projected activities for the design basis reference spent nuclear fuel, and the annual limit of ingestion set by the Nuclear Safety and Security Commission Notification No. 2019-10 without considering their chemical and hydrogeological properties. The proposed process was used to select 56 radionuclides, comprising 27 fission and activation products and 29 actinide nuclides. This study explains first the determination of the design basis reference spent nuclear fuels, followed by a comprehensive discussion on the selection criteria and methodology for safety-relevant radionuclides.

Keywords: Deep geological repository, Long-term safety assessment, Safety-relevant radionuclide, Design basis reference spent nuclear fuel, Source terms, SCALE 6.2.4

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1. Introduction

Deep geological repository refers to the long-term isolation of radioactive waste in stable deep rock formations. This concept employs a multi-barrier system composed of engineering and natural barriers [1]. Initially proposed in the 1950s in the United States [2], six decades of technological advancements have made deep geological repository the most widely accepted ultimate management solution for HLW (High-Level Waste) and spent nuclear fuel in countries operating nuclear power plants [3]. Some nations even classify ILW (Intermediate-Level Waste) as suitable for deep geological repositories. In recent years, significant progress has been made, particularly in Europe. Finland is constructing the world's first deep disposal repository of spent nuclear fuel [4]. Sweden obtained a construction license in 2022, and France has selected a site for its deep disposal repository and aims to submit its construction license by January 2023. Other countries like Switzerland, China, and Russia are also making significant strides in this area [5].

As of March 31, 2023, South Korea operates 25 nuclear reactors, comprising 22 PWRs (Pressurized Water Reactor) and three PHWRs (Pressurized Heavy Water Reactor), with a total installed capacity of 24.7 GWe. Furthermore, based on the 10th Basic Plan for Long-term Electricity Supply and Demand of South Korea, the country plans to expand its nuclear capacity to 31.7 GWe by 2036 through the construction of new PWRs such as Shin Hanwol Units 2, 3, 4, and Shin Kori Units 5 and 6. This expansion is estimated to generate a total of 72,035 bundles of PWR spent nuclear fuel. It is predicted that the spent nuclear fuel storage pools at the Hanbit, Hanul, and Kori sites will reach saturation by 2030, 2031, and 2032, respectively [6].

Under these situations, spent nuclear fuel management has emerged as a high-priority issue for South Korea's long-term energy sustainability, necessitating rigorous action and technology development. In response, the government of South Korea has led a comprehensive, long-term program to develop the essential technologies for the deep geological repository of spent nuclear fuels. KORAD (Korea Radioactive Waste Agency) is at the forefront of these efforts, initiating an extensive research and development project to enhance technical competencies for managing high-level radioactive waste in a deep geological repository. One of the project's primary objectives is to evaluate the long-term safety and performance of such a disposal system. This involves understanding the complex, coupled processes of thermal (T), hydraulic (H), mechanical (M), and chemical (C) mechanisms, which are integral to the repository's integrity. A fundamental step in this evaluation is the source term assessment, which requires an in-depth analysis of the characteristics of the spent nuclear fuels.

The objective of this work is to establish a screening procedure for selecting safety-relevant radionuclides within the framework of long-term safety assessments for deep geological repositories to provide the important nuclides for the initial consideration in the long-term safety assessments. However, it is important to note that the criteria for the selection of radionuclides in this study are based on the projected activity and half-lives of the radionuclides to be disposed of in the repository. The chemical and hydrogeology properties of the radionuclides have not been considered. Additionally, this work has conducted statistical analysis on the characteristics of domestic spent nuclear fuels to select the design basis reference spent nuclear fuels. Also, we evaluated the portion of the activity and decay heat from the selected safety-relevant radionuclides in the design basis reference spent nuclear fuel for up to one million years.

The rest of this paper is organized as follows: Section 2 provides the design basis reference spent nuclear fuels, with subsections 2.1 reviewing the statistical analysis and determination of PWR spent nuclear fuels. Section 2.2 presents the results of the design basis reference spent nuclear fuel source term calculation. Section 3 is dedicated to the selection of safety-relevant radionuclides, with subsection 3.1 reviewing other country procedures, 3.2 outlining the procedure and 3.2 discussing the selected radionuclides. Finally, Section 4 offers the conclusion.

2. Design Basis Reference Spent Nuclear Fuel Assemblies

2.1 Statistical Analysis of PWR Spent Nuclear Fuels

In this section, the determination of the design basis reference spent nuclear fuels is reviewed. The determination of the design basis reference spent nuclear fuels was performed through the statistical analysis using the Korea PWR spent nuclear fuels database from Korea Hydro & Nuclear Power (KHNP), which was provided through the Korea Radioactive Waste Agency (KORAD). This database contains 20,970 spent nuclear fuels discharged after 1979 up to 2021. The database includes detailed information about the discharged spent nuclear fuels and the associated reactor data. For each assembly, the database contains the initial uranium mass, the ^{235}U enrichment, the discharge burnup, the last cycle discharge date, the specific power, the assembly type, and the effective full power day [7].

The spent nuclear fuels in South Korea are categorized into four main lattice types: WH 14×14, WH 16×16, WH 17×17, and CE 16×16. These lattice types further branch into various subtypes based on slight variations in uranium loadings despite having identical lattice specifications. In terms of portion, WH 17×17 and CE 16×16 lattice types dominate, comprising 42.6% and 43.5% of the total spent nuclear fuel assemblies, respectively. The WH 14×14 and WH 16×16 types comprise smaller portions, at 6.6% and 7.3%.

Fig. 1 shows the number of spent nuclear fuel assemblies in South Korea for initial enrichments and discharge burnups (as of December 2021). Most spent nuclear fuels exhibit discharge burnups ranging between 25 and 55 MWd·kg^{-1}. It is noted that 70% of all assemblies have burnups lower than 45 MWd·kg^{-1}. Consequently, we categorize spent nuclear fuel assemblies into two groups using a 45 MWd·kg^{-1} cut-off burnup point. The first group consists of assemblies with a burnup value less than 45 MWd·kg^{-1},

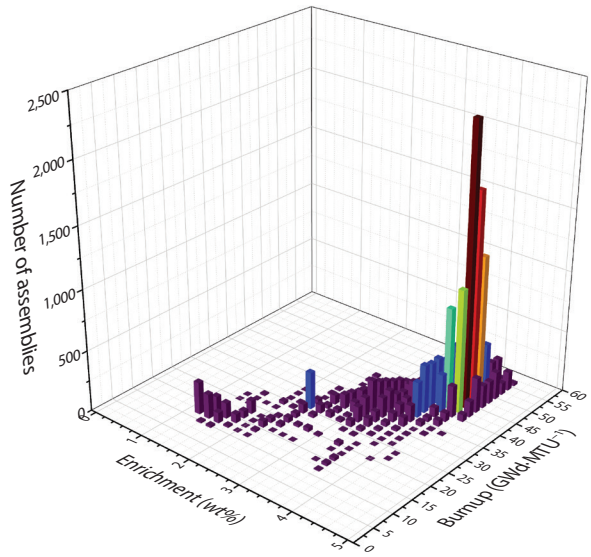

Fig. 1. Distribution of initial enrichment and discharge burnups of spent nuclear fuels.

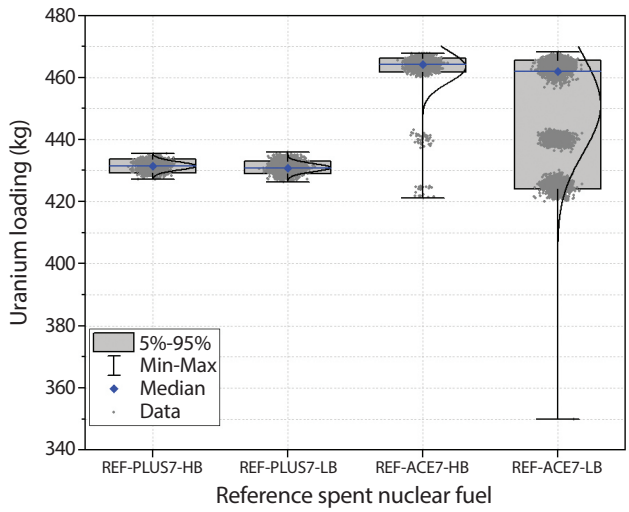

Fig. 2. Statistical values of uranium loading for low and high burnup groups of spent nuclear fuel assemblies.

whereas the second group includes those with a burnup value exceeding 45 MWd·kg^{-1}. The representative burnup values for the low and high burnup groups are set to 45 MWd·kg^{-1} and 55 MWd·kg^{-1}, respectively.

Recent trends indicate increases both in initial uranium enrichments and burnups, especially for long-cycle operations. By December 2021, the inventory included

Table 1. Specification of the design basis reference spent nuclear fuels

Name	Low burnup group		High burnup group	
	REF-PLUS7-LB	REF-ACE7-LB	REF-PLUS7-HB	REF-ACE7-HB
Lattice type	16×16	17×17	16×16	17×17
Burnup ($MWd \cdot kg^{-1}$)	45	45	55	55
Initial uranium enrichment (%)	4.5	4.5	4.65	4.65
Initial uranium loading (kg)	436	468	436	468
Specific power ($MW \cdot MTU^{-1}$)	40	40	40	40
Depletion interval (EFPDs)	1,125	1,125	1,375	1,375

approximately 2,347 spent nuclear fuel assemblies with uranium enrichments exceeding 4.6%. To maintain a conservative approach, the design basis reference spent nuclear fuel assemblies are designated with initial uranium enrichments of 4.5% for the low burnup group and 4.65% for the high burnup group. Fig. 2 shows the statistics of the initial uranium loadings for low and high burnup groups of the CE 16×16 (designated as PLUS 7 in Fig. 2) and WH 17×17 (designated as ACE 7 in Fig. 2) assemblies. The statistics of the initial uranium loadings indicate maximum uranium loadings of 435,984 grams and 435,581 grams for the low and high burnup groups of the CE 16×16 assemblies, respectively. While the corresponding values for the WH 17×17 assemblies are 468,259 grams and 467,770 grams. Consequently, the initial uranium loadings for CE 16×16 and WH 17×17 types are set to be 436 kg and 468 kg, irrespective of their burnup groups. Table 1 summarizes the specification of the design basis reference spent nuclear fuels.

2.2 Source Term of Design Basis Reference Spent Nuclear Fuels

In this work, we used SCALE 6.2.4 to calculate the source terms for the design basis reference spent nuclear fuels. We utilized its SCALE 6.2.4/TRITON module to produce burnup-dependent neutron cross section libraries for standalone ORIGEN calculations. SCALE 6.2.4/ORIGEN is a stand-alone depletion code, which calculates

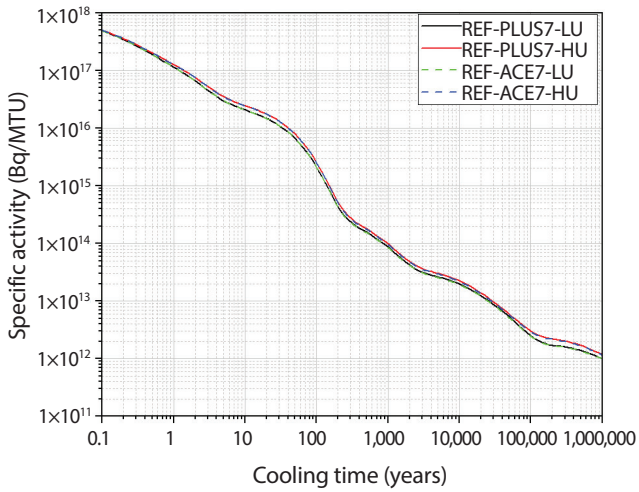

Fig. 3. Specific activity as a function of cooling time for each group.

time-dependent concentrations, activities, and radiation source terms for a large number of isotopes simultaneously generated or depleted by neutron transmutation, fission, and radioactive decay. TRITON solves multi-group transport equations coupled with depletion calculations and allows users to save assembly-average neutron cross sections for future ORIGEN use [8]. We ran multiple TRITON simulations to cover parameter variations like uranium enrichment and moderator density. These provided a comprehensive set of ORIGEN cross section libraries.

Calculations for burnup and cooling were performed for the design basis reference spent nuclear fuels, ranging from the time of discharge from the reactor up to 1,000,000 years. Evaluation results include calculations for activity,

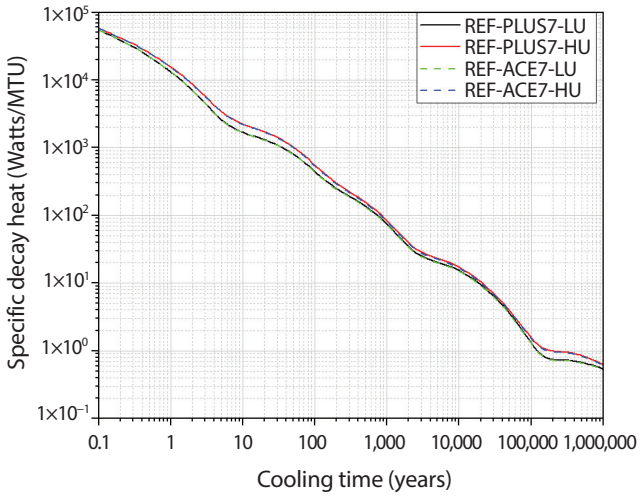

Fig. 4. Specific decay heat as a function of cooling time for each group.

decay heat, nuclide inventory, and neutron/gamma spectra. All evaluation results were normalized per metric ton of uranium (MTU). These results can be converted to per assembly results by multiplying with the initial uranium loading for the given fuel assembly. For example, Figs. 3 and 4 show the total specific activity and specific decay heat of each design basis reference spent nuclear fuels, respectively. Especially, the decay heat is one of the most important parameters in analyzing the long-term safety assessment for deep geological repository.

3. Selection of Safety-relevant Radionuclides

In the long-term safety assessment of deep geological repository, it is anticipated that radionuclides may be released through engineered barriers due to functional defects in spent nuclear fuel or disposal canisters. Spent nuclear fuel contains many radionuclides, requiring dose assessment upon their potential release into the ecosystem. The total number of spent nuclear fuel assemblies for the geological disposal repository was estimated 72,036 as stated in the 10th Basic Plan for Long-term Electricity Supply and Demand of South Korea. REF-ACE7-HB has an initial uranium mass of 468 kg, therefore the total initial uranium mass of 72,036 spent nuclear fuel assemblies is 33,712 MTU. In Section 2, we first calculated the total activity (Bq) and the nuclide-wise activity (Bq) per one metric ton uranium (1 MTU) for the selected design basis reference spent nuclear fuel over the cooling period. So, we calculated the activity of each radionuclide present in one assembly of spent nuclear fuel by multiplying it by 0.468 MTU (i.e., 468 kg) with the activity per 1 MTU for the design basis reference spent nuclear fuel. Then, we estimated the total nuclide-wise radioactivity for the 72,036 spent nuclear fuel assemblies (equivalent to 33,712 MTU). Then, these nuclide-wise activity are used both in Steps 2 and 3 of Section 3.2. The REF-ACE7-HB was used due to its high initial uranium loading and burnup, which generally results in higher inventories of the radionuclides.

Additionally, prior to conducting an actual ecosystem assessment, highly conservative assumptions were made to select safety-relevant isotopes. The physical half-life of the radionuclide and the ALI (Annual Limit of Ingestion) set by the NSSC (Nuclear Safety and Security Commission) Notification No. 2019-10 were used as criteria for selection. This study's selection procedure does not consider the isotope delay effects of the engineered and natural barriers of the disposal system; therefore, chemical properties such as solubility limits in groundwater, backfill materials, and distribution coefficients for buffering materials were not included. So, some of these nuclides can be removed in a specific analysis area depending on their properties.

Given the constraints of basing evaluations on the design basis reference spent nuclear fuel source terms, it is impractical to consider every radionuclide or their respective radionuclides that may contribute a significant radiation dose, as well as all potential transport pathways from the repository to the ecosystem. In this way, isotopes that do not contribute substantially to overall risk were excluded, and conservative assumptions about isotope transport pathways were made.

3.1 Review of Safety-relevant Radionuclides Selection Procedure

In this subsection, the selections of the safety-relevant nuclides from other institutes are briefly reviewed. First, POSIVA used the four steps for this purpose [9]. In the first step, 1,284 nuclides having low specific activity of less than 10^{-30} GBq/MTU were excluded, and 201 nuclides were selected. In the second step, the RQs (Risk Quotient) for nuclides were used to exclude some nuclides. The RQ is defined as a normalized product of the maximum activity of a nuclide overall time and the dose conversion factor for the ingestion of the same nuclide. In the second step, the nuclides having small RQ values less than 1.0 were excluded. Also, the short-lived nuclides having shorter half-lives than 1 year were excluded in the step 3, which gave 84 nuclides. In the fourth step, 41 nuclides were selected with some additional considerations such as solubility limits, ingestion dose index, and short-lived daughter nuclides in the decay chains, and IRF nuclides.

SKB classified the nuclides into the fission product nuclides, activation nuclides, and the nuclides in the decay chains for screening purpose [10]. For the fission product and activation nuclides, SKB used two steps for screening. In the first step, the nuclides that have shorter half-lives than 10 years and lower radiotoxicity index (RTI) less than 0.01 were excluded. The radiotoxicity index was defined as the hypothetical dose at a time resulting from ingestion of the activity A_j [Bq], divided by 10^{-4} Sv, which is given by

$$RTI(t) = \frac{\sum_j A_j(t) D_j}{10^{-4} Sv} \quad (1)$$

where D_j [Sv·Bq^{-1}] is the dose coefficient for ingestion.

Then, in the second step, 23 nuclides selected in the first step are divided into 10 less-important nuclides and 13 important nuclides, depending on radiotoxicity, half-life, and annual dose. For the decay chain nuclides, SKB used three steps for screening. In the first step, the shorter-lived nuclides than 1 year were excluded and the nuclides having almost zero inventories were excluded in the second step. In the third step, the nuclides of which the annual dose significantly decreases before 1,000 years after disclosure of the geological repository were excluded, which led to 22 nuclides. So, SKB finally selected 45 nuclides (i.e., 23 fission product and activation nuclides, and 22 decay chain nuclides).

Nagra also used the same classification of the nuclides (i.e., classification into fission product and activation nuclides, and decay chain nuclides) [11]. They excluded the nuclides having the annual water consumption of a representative person less than 10^{-5} mSv·a^{-1} but considered some indicator nuclides (i.e., ^{3}H, ^{90}Sr, and ^{137}Cs) for phenomena that occur at relatively early times after the end of emplacement (e.g., pinhole release for defective SF/HLW canisters). The screening criterion based on the annual water consumption is similar to the equation (i.e., Eq. (3)) we used in this work. Also, they excluded six daughter nuclides (i.e., ^{90}Y, ^{121}Sn, ^{126m}Sb, ^{210}Bi, ^{214}Pb, and ^{214}Bi) having short half-lives by adding their dose coefficients to the longer-lived parent radionuclide. Through these screenings, they selected 22 fission products and activation nuclides. For the decay chain nuclides, 29 nuclides were selected by excluding 43 daughter nuclides having short half-lives by adding their dose coefficients to the longer-lived parent radionuclide. Finally, they considered 22 fission products and activation nuclides, and 29 decay chain nuclides.

3.2 The Procedure of the Safety-relevant Radionuclides Selection

The selection procedure used in this work is given in Fig. 5.

The procedure can be explained by the following four steps:

Step 1: Out of numerous radionuclides, 1,417 were initially selected, excluding stable isotopes.

Step 2: 254 radionuclides with a specific activity exceeding 10^{-30} Bq/MTU were selected.

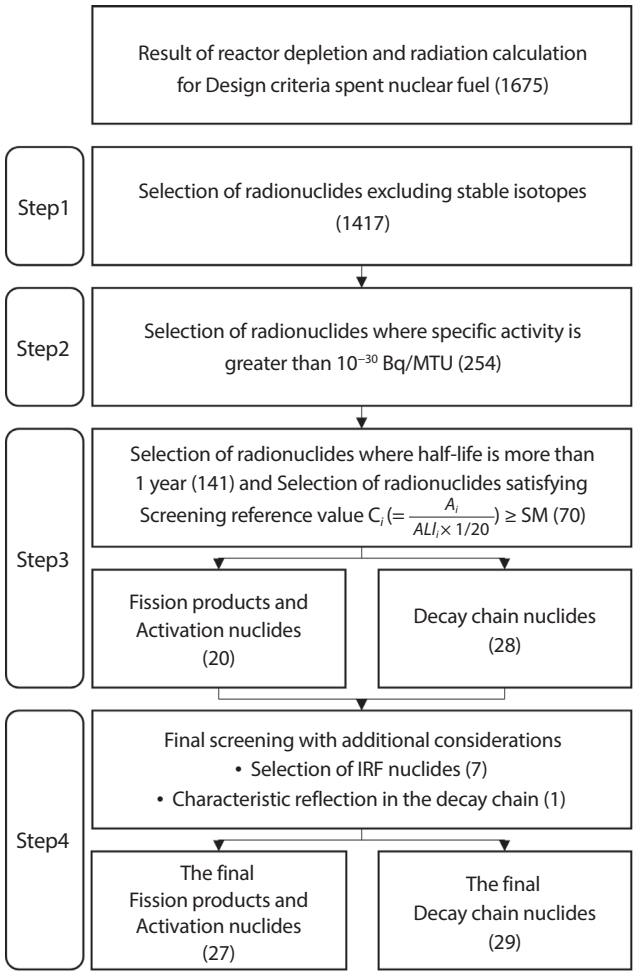

Fig. 5. The schematic of the safety-relevant radionuclides selection.

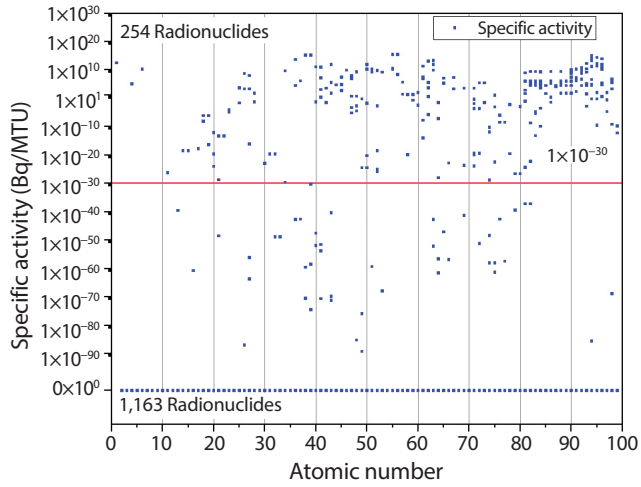

Fig. 6. Radionuclides screened in Step 2.

Fig. 6 shows the atomic numbers for 1,417 radionuclides selected in Step 1, screening them into two categories: 1) 1,163 radionuclides with specific activity below 10^{-30} Bq/MTU and 2) 254 radionuclides exceeding this level.

Step 3: A total of 48 radionuclides with a half-life of more than 1 year and satisfying the Selection Criterion Index (C_i) were selected. The Selection Criterion Index (C_i) was defined as:

$$C_i = \frac{A_i}{ALI_i \times 1/20} \geq SM \qquad (2)$$

where, A_i is the annual ingestion of radionuclide i [Bq·yr^{-1}], ALI_i is the Annual Limit of Ingestion of radionuclide i [Bq·yr^{-1}], and SM is the safety margin, 10^{-3}.

At this stage, the defined selection criteria do not consider the configuration, features, detailed site-specific groundwater flow, and geochemical models, nor detailed dose assessments in ecosystems. Radionuclides were selected based on the annual ingestion (A_i), assuming that the public would drink the water released from spent nuclear fuel according to the design criteria and that all the radionuclides were dissolved into the water. The Annual Limit of Ingestion (ALI_i) for radionuclide, as specified by the NSSC (Nuclear Safety and Security Commission) Notification No. 2019-10, is based on the dose limit for workers (20 mSv·yr^{-1}). In this work, the standard value was reduced by multiplying the ALI_i by 1/20 to match the public dose limit (1 mSv·yr^{-1}). Then, the radionuclides were selected by considering the safety margin (SM) of 10^{-3} to the reduced ALI_i. Actually, the safety margin of 10^{-3} corresponds to the annual dose limit of 1 μSv·yr^{-1}. However, it is needed to estimate the annual ingestion (A_i) of a radionuclide for applying the criteria given in Eq. (2). In this work, the annual ingestion was estimated using the following assumptions:

- After the designed lifetime of the disposal canister, all isotopes are assumed to dissolve the Quaternary aquifers.

- No consideration was given to the solubility limits of isotopes.
- The aquifer is assumed to be consumed as drinking water.
- No account was taken of the effects of adsorption, retardation, and accumulation due to the engineered and natural barriers of deep geological repositories.

Given these considerations, the annual ingestion (A_i) of radionuclides *i* was calculated using the following equation:

$$A_i = \frac{N_i^{max} \cdot I_{wat}}{T_c \cdot Q} \quad (3)$$

where, N_i^{max} represents the maximum activity of a radionuclide *i* over the design lifetime of the disposal canister [Bq]. I_{wat} denotes the annual water consumption of a representative person [$m^3 \cdot yr^{-1}$]. T_c is the effect time of general release through the near-field [yr]. Q is the flow rate of the aquifer [$m^3 \cdot yr^{-1}$]. In other word, the maximum activity of the considered nuclide is dilluted into the Quaternary aquifers of which the amount is determined by multiplying the flow rate with the generic duration of near-field release, and then the water flows to the surface where human drinks the water [11].

In this work, the total activity of a radionuclide was determined by multiplying the total amount of waste in the deep disposal repository by the activity per metric ton of uranium (Bq/MTU) for the domestic design basis reference spent nuclear fuels. Also, it was assumed that four bundles of design basis reference spent nuclear fuel (1.872 MTU) would be disposed of in each disposal canister. According to the 10th Basic Plan for Long-term Electricity Supply and Demand of South Korea, there will be 18,009 disposal canisters (equivalent to 72,036 bundles of spent nuclear fuel). The effective time of general release through the near-field is assumed to be 10,000 years. The maximum activity of each radionuclide between 40 years and 10,000 years after its discharge from the reactor for the design basis reference spent nuclear fuels has been used for calculations. The annual water consumption of a representative person is assumed to be 0.73 $m^3 \cdot yr^{-1}$ [12]. The flow rate of the aquifer is assumed to be 10^6 $m^3 \cdot yr^{-1}$ based on the international literature, specifically from Swiss Nagra and Japanese JNC studies [11, 13], since a domestic disposal site has not yet been determined. An increase in the flow rate of the aquifer and the design lifetime of the disposal canister implies a decrease in the concentration of dissolved radionuclides, which leads to a decrease in the nuclides to be considered. In the second stage of the selection process, 254 radionuclides were considered. 141 have a half-life greater than one year. 70 radionuclides meet the Selection Criterion Index (C_i), and 48 radionuclides meet both criteria.

Step 4: The radionuclides selected in Step 3 were classified into fission and activation products and decay chain nuclides. 20 fission and activation products were selected in Step 3. Although some were excluded in Step 3 because they did not meet the selection criteria, seven additional nuclides (i.e., ^{108m}Ag, ^{36}Cl, ^{93}Mo, ^{94}Nb, ^{59}Ni, ^{63}Ni, and ^{107}Pd) were included because they are considered as the IRF (Instant Release Fraction) nuclides. As a result, 27 fission products and activation nuclides were selected. Table 2 lists the IRF radionuclides selected from the studies of POSVIA and SKB, all of which were included in the nuclide selection process of this work [9, 10]. Radionuclides release rapidly with contacting water or slowly with the dissolution of the UO_2 matrix. The former is known as the 'Instant Release', and the latter is 'Congruential Release'. Even though the IRF are much smaller than the congruential ones, IRF has to be treated carefully due to the fact that the instant releases lead to a much larger value of the exposure dose rates than the congruential ones which proceed very slowly.

On the other hand, the actinide nuclides exist in four groups (i.e., 4N, 4N+1, 4N+2, and 4N+3) of alpha decay, with no interconnections between the different groups. Typically, daughter nuclides with much shorter half-lives than their parent nuclides are assumed to reach equilibrium, simplifying the decay chain. Furthermore, parent nuclides with shorter physical half-lives are incorporated into the initial

Table 2. List of IRF radionuclides selected by POSIVA and SKB

POSIVA	SKB
^{108m}Ag	^{108m}Ag
^{14}C	^{14}C
	^{113m}Cd
^{36}Cl	^{36}Cl
^{135}Cs	^{135}Cs
^{137}Cs	^{137}Cs
	^{3}H
^{129}I	^{129}I
^{93}Mo	^{93}Mo
	^{93m}Nb
	^{94}Nb
	^{59}Ni
	^{63}Ni
^{107}Pd	^{107}Pd
^{79}Se	^{79}Se
	^{121m}Sn
^{126}Sn	^{126}Sn
^{90}Sr	^{90}Sr
^{99}Tc	^{99}Tc
	^{93}Zr

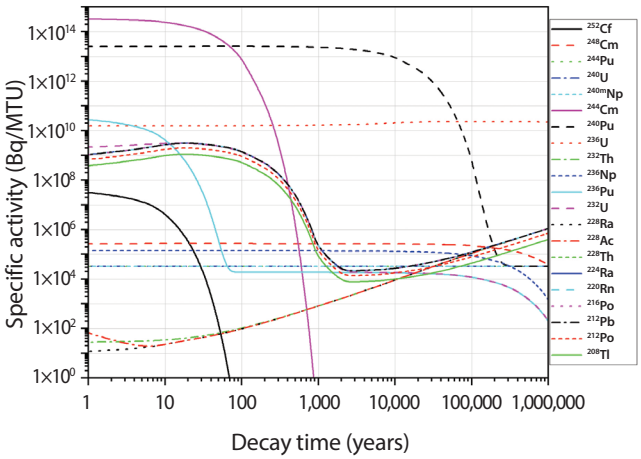

Fig. 7. Specific activity (Bq/MTU) as a function of decay time for radionuclides in the 4N decay chain.

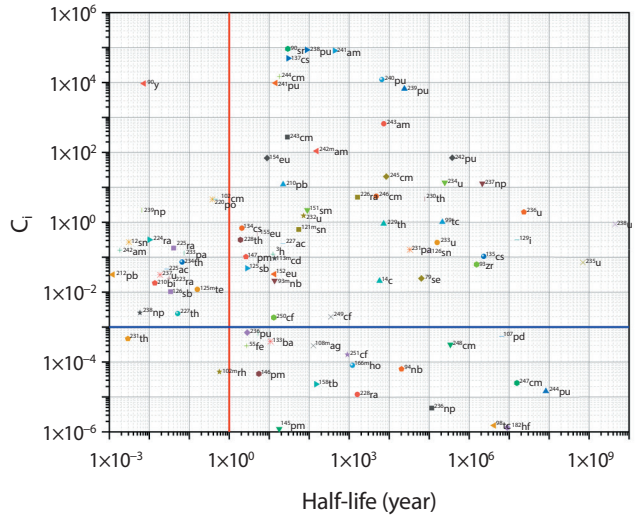

Fig. 8. Radionuclides selected for safety-relevant in the Step 3.

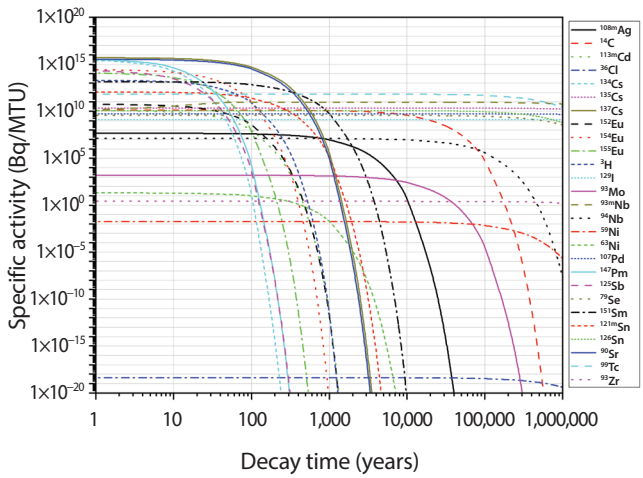

Fig. 9. Specific activity (Bq/MTU) as a function of decay time for the selected radionuclides in fission and activation products.

activity of their daughter nuclides to simplify the decay chain. However, their impact on activity is still included in subsequent dose assessments [10]. These considerations and using the selection criteria concerning half-life and C_i led to the selection of 28 actinide nuclides.

However, ^{232}Th, which is a part of the 4N decay chain, was additionally included because its inventory steadily increases and reaches equilibrium with its daughter nuclides, as shown in Fig. 7. Fig. 7 shows the changes of the

Table 3. List of the safety-relevant radionuclides from fission and activation products

Radionuclides	Half-life (year)	Satisfy the C_i	IRF
^{3}H	1.23×10^{1}	Satisfy	IRF
^{14}C	5.70×10^{3}	Satisfy	IRF
^{36}Cl	3.01×10^{5}	-	IRF
^{59}Ni	7.60×10^{4}	-	IRF
^{63}Ni	1.01×10^{2}	-	IRF
^{79}Se	2.95×10^{5}	Satisfy	IRF
^{90}Sr	2.88×10^{1}	Satisfy	IRF
^{93}Zr	1.53×10^{6}	Satisfy	IRF
^{93m}Nb	1.61×10^{1}	Satisfy	IRF
^{94}Nb	2.03×10^{4}	-	IRF
^{93}Mo	4.00×10^{3}	-	IRF
^{99}Tc	2.11×10^{5}	Satisfy	IRF
^{107}Pd	6.50×10^{6}	-	IRF
^{108m}Ag	4.38×10^{2}	-	IRF
^{113m}Cd	1.41×10^{1}	Satisfy	IRF
^{121m}Sn	4.39×10^{1}	Satisfy	IRF
^{126}Sn	2.30×10^{5}	Satisfy	IRF
^{125}Sb	2.76×10^{0}	Satisfy	-
^{129}I	1.57×10^{7}	Satisfy	IRF
^{134}Cs	2.07×10^{0}	Satisfy	-
^{135}Cs	2.30×10^{6}	Satisfy	IRF
^{137}Cs	3.01×10^{1}	Satisfy	IRF
^{147}Pm	2.62×10^{0}	Satisfy	-
^{151}Sm	9.00×10^{1}	Satisfy	-
^{152}Eu	1.35×10^{1}	Satisfy	-
^{154}Eu	8.60×10^{0}	Satisfy	-
^{155}Eu	4.75×10^{0}	Satisfy	-

Table 4. List of the safety-relevant radionuclides from the decay chains

Decay chain	Radionuclides	Half-life (year)
4N	^{244}Cm	1.8110×10^{1}
	^{240}Pu	6.5610×10^{3}
	^{236}U	2.3420×10^{7}
	^{232}Th	1.4050×10^{10}
	^{232}U	6.8900×10^{1}
	^{228}Th	1.9120×10^{0}
4N+1	^{249}Cf	3.5100×10^{2}
	^{245}Cm	8.5000×10^{3}
	^{241}Pu	1.4290×10^{1}
	^{241}Am	4.3260×10^{2}
	^{237}Np	2.1440×10^{6}
	^{233}U	1.5920×10^{5}
	^{229}Th	7.3400×10^{3}
4N+2	^{250}Cf	1.3080×10^{1}
	^{246}Cm	4.7678×10^{3}
	^{242m}Am	1.4100×10^{2}
	^{242}Pu	3.7350×10^{5}
	^{238}U	4.4680×10^{9}
	^{238}Pu	8.7700×10^{1}
	^{234}U	2.4550×10^{5}
	^{230}Th	7.5380×10^{4}
	^{226}Ra	1.6000×10^{3}
	^{210}Pb	2.2200×10^{1}
4N+3	^{243}Am	7.3700×10^{3}
	^{243}Cm	2.9100×10^{1}
	^{239}Pu	2.4110×10^{4}
	^{235}U	7.0380×10^{8}
	^{231}Pa	3.2760×10^{4}
	^{227}Ac	2.1772×10^{1}

activities of the nuclides in the 4N decay chain. As a result, 29 actinide nuclides were selected through the third stage. This selection process ultimately yielded a total of 56 selected radionuclides. Fig. 8 shows the radionuclides selected based on the criteria of the Step 3, categorizing them by their half-lives and the Selection Criterion Index (C_i). The data for the half-lives was used from the ENDF/B-VII.1 nuclear data library used in ORIGEN [14].

Table 3 represents the list of safety-relevant radionuclides from fission and activation products that were finally selected in Step 4 based on the Selection Criterion Index (C_i), a half-life greater than one year, and their classification as the IRF nuclides. Fig. 9 shows the changes in the selected activities of the fission product and activation products.

Table 4 shows the selected safety-relevant radionuclides

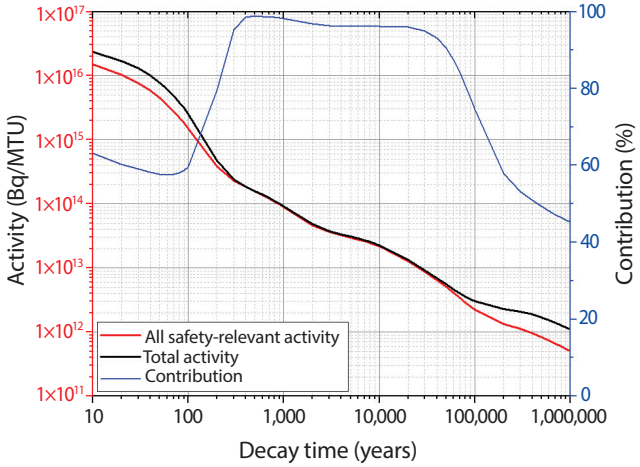

Fig. 10. Contribution of safety-relevant radionuclides to total activity.

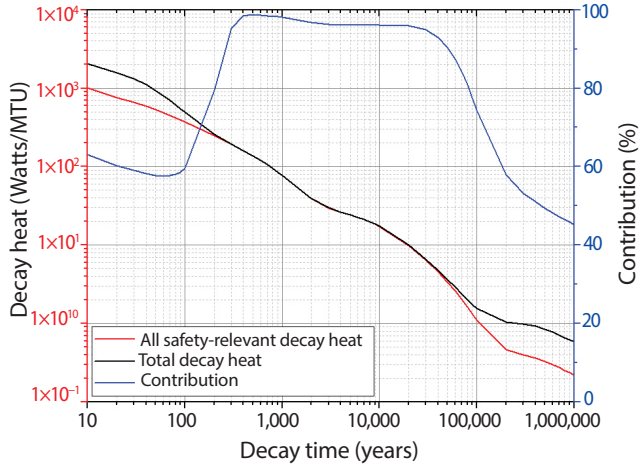

Fig. 11. Contribution of safety-relevant radionuclides to total decay heat.

in the 4N, 4N+1, 4N+2, and 4N+3 decay chains that were finally selected in Step 4 based on the Selection Criterion Index (C_i), a half-life of greater than one year.

3.3 Analysis for Safety-relevant Radionuclides

Figs. 10 and 11 show the contribution of the finally selected safety-relevant radionuclides to total activity and total decay heat, respectively. The contribution of safety-relevant radionuclides to the total activity and decay heat of spent nuclear fuel is approximately 60% up to 300 years after discharge from the reactor. The remaining contribution of ~40% is largely attributed to the short half-life radionuclides such as ^{137m}Ba (2.55 min), ^{90}Y (64.2 h), ^{239}Np (2.36 d), ^{121}Sn (27 h), and ^{126m}Sb (11 s). Once these radionuclides have fully decayed, the contribution of the safety-relevant radionuclides to total activity rises to over 90%. The contribution of safety-relevant radionuclides to the total activity decreases after 100,000 years. This decline can be attributed to the influence of daughter radionuclides with shorter half-lives reaching equilibrium with their parent radionuclides in each decay chain. In the 4N decay chain, the daughter radionuclides such as ^{228}Ac, ^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Po, and ^{208}Tl are in equilibrium with ^{232}Th. In the 4N+1 decay chain, the daughter radionuclides such as ^{225}Ra, ^{225}Ac, ^{221}Fr, ^{217}At, ^{213}Bi, ^{213}Po, and ^{209}Tl also are in equilibrium with ^{229}Th. In the 4N+2 decay chain, the daughter radionuclides such as ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po, ^{210}Bi, and ^{210}Po are in equilibrium with ^{226}Ra. Lastly, in the 4N+3 decay chain, the daughter radionuclides such as ^{227}Th, ^{223}Fr, ^{223}Ra, ^{219}Rn, ^{215}Po, ^{211}Pb, ^{211}Bi, ^{211}Po, and ^{207}Tl reach an equilibrium state with ^{227}Ac. Therefore, when performing dose assessments, the contributions of these daughter radionuclides are considered in the initial inventory of the parent radionuclides.

4. Conclusion

In this work, a procedure for selecting the safety-relevant radionuclides was established based on the criteria in terms of the half-lives, the ALI set by the NSSC (Nuclear Safety and Security Commission) Notification No. 2019-10, and some additional considerations such as the inclusion of IRF nuclides. Before the establishment of the procedure, the design basis reference spent nuclear fuels were determined through a statistical analysis based on the spent nuclear fuel database in South Korea. In particular, the design basis reference spent nuclear fuel assemblies were determined for each of the WH 17×17 and CE 16×16 assembly types and

low and high burnup groups. For the CE 16×16 type, the PLUS 7 spent nuclear fuel assembly having 436 kg initial uranium loading was determined as the design basis reference spent nuclear fuel, while the ACE 7 spent nuclear fuel assembly having 468 kg initial uranium loading was selected for the WH 17×17 type. The representative burnups for low and high burnup groups were determined to be 45 and 55 $MWd \cdot kg^{-1}$, respectively, and the initial uranium enrichments for these burnup groups were determined to be 4.5 and 4.65wt%, respectively, irrespective of the assembly types. The nuclide-wise activities for the design basis reference spent nuclear fuels over 1,000,000 years after discharge were determined using the ORIGEN code in SCALE 6.2.4 for use in the selection procedure of the safety-relevant nuclides.

In the criteria related to ALI, a conservative safety margin of 10^{-3} corresponding to the annual dose limit of 1 $\mu Sv \cdot yr^{-1}$ was used coupled with some simple and conservative assumptions in determining the nuclide-wise annual ingestion. These assumptions include 1) no consideration of solubility limits, 2) no consideration of the effects of adsorption, retardation, and accumulation due to the engineered and natural barriers of deep geological repositories, 3) full dissolution of the radionuclides in the drinking water near the ground surface aquifers, and 4) use of maximum nuclide-wise activity over the design lifetime of the canister. The maximum nuclide-wise activity was computed for the design basis reference spent nuclear fuels of high burnup based on the 10th Basic Plan for Long-term Electricity Supply and Demand of South Korea.

As the results of the analysis, we selected a total of 27 fission product and activation product nuclides (^{108m}Ag, ^{36}Cl, ^{93}Mo, ^{94}Nb, ^{59}Ni, ^{63}Ni, and ^{107}Pd were included due to their instant release characteristics even if they did not satisfy the selection criteria) and 29 decay chain nuclides, giving totally 56 radionuclides as the safety-relevant nuclides. Also, from the analysis of the contributions of these nuclides, it was found that their contribution to total activity and decay heat range from 50–70% over the initial stage of about 100 years after discharge while their contribution is very high (78–98%) over 300–100,000 years after discharge. Finally, it should be noted that the selected safety-relevant nuclides in this work can be used at the initial stage of the safety assessments of the deep geological disposal system, but some of them can be excluded in the later, more sophisticated stage depending on the safety assessment area.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Acknowledgements

This work was supported by the Institute for Korea Spent Nuclear Fuel (iKSNF) and Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government (Ministry of Trade, Industry and Energy (MOTIE)) (No. 2021040101003C).

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