Selection of Reference PWR Spent Nuclear Fuel and Source Term Calculation for Deep Geological Disposal System

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1. Introduction

As of the third quarter of 2021, 504,809 assemblies of spent nuclear fuel (SNF) were generated (i.e., 20,733 PWR assemblies and 484,076 PHWR assemblies) in our country. The on-site storage facilities are expected to be saturated sequentially from 2031 due to the continuous generation of SNFs [1]. Therefore, it is urgent and important to construct a geological repository to dispose of the SNFs.

The objective of this work is to select the design basis reference SNF and to evaluate the source term of the SNF to establish the evaluation technology for the deep geological SNF repository and the safety verification system. Based on the domestic SNFs generation database up to December 2021, statistical analysis was performed to select the design basis reference SNF considering the fuel assembly specifications, cooling time, and burnups. Using the SCALE6.2.4/ORIGEN-ARP computer code, the source term evaluation was performed on the design basis reference SNF. The results were made into a database (e.g., radioactivity, decay heat, gamma heat, plutonium contents, neutron/gamma spectrum, and nuclide inventory), which will be utilized in the future study of evaluation of the performances of the national deep geological disposal system.

2. Methods and Results

It was possible to refer to the Korea PWR SNFs database from Korea Hydro & Nuclear Power (KHNP) through the Korea Radioactive Waste Agency (KORAD). This database contains 20,970 SNF discharged after 1979 up to 2021. The database includes detailed information about the discharged SNFs 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.

2.1 Statistics and Distribution of SNFs Properties

This section describes the database-based statistics and distribution of SNFs properties. Fig. 1 shows the number of annual SNFs as a function of the discharge year. In addition, the cumulative percentage is also shown. As shown, the number of discharged SNFs increased significantly from the late 1980s and early 2000s as more reactors came into service. It can be shown that since 2005 (excluding 2020), 750 SNFs on the average have been discharged every year.

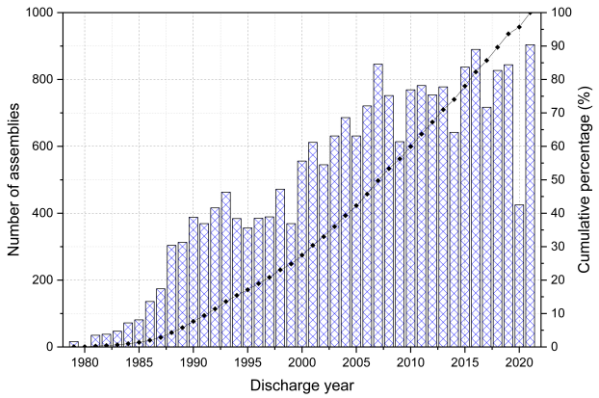

Fig. 1. Annual number of fuel assemblies discharged

Fig. 2 shows the number of SNFs as a function of fuel burnup. The x-axis of the histogram represents the discharged burnup of SNFs, and each bin in burnup is 1 MWd/kg. As shown, the burnup varies within 23 ~ 55 MWd/kg. Average and median burnups are estimated to be 40 MWd/kg and 42 MWd/kg, respectively.

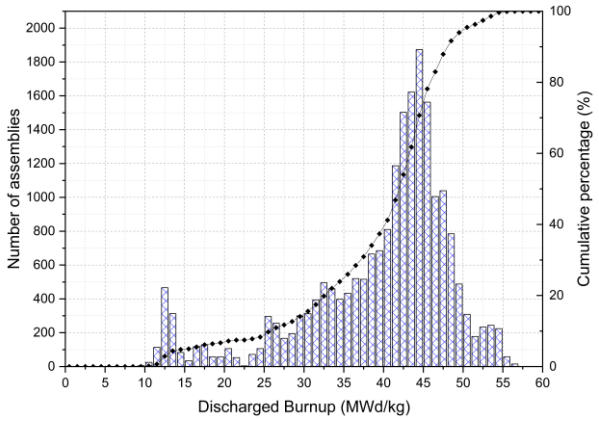

Fig. 2. Number of assemblies as a function of discharged burnup (x-axis bin: 1 MWd/kg)

The initial fuel enrichment closely correlates with the burnup of the SNFs. Fig. 3 represents the change of the median initial fuel enrichment as a function of discharge year for assemblies. In Fig. 3, the 5 % and 95 % enrichment values are shown to illustrate the range of uranium enrichment from a minimum up to a maximum of the assemblies. The figure shows that the median enrichment increases with each passing year, except

from 1979 to 1988, when many new reactors started their operation for the first time. The development of the second-generation fuels (e.g., ACE7, PLUS7) with the increased allowable burnup and thermal performances enhances fuel integrity, economics, and reliability compared to the first-generation fuels, which explains a significant amount of loading of nuclear fuels with enrichment of 4.0 wt% or more since the 2000s [2, 3]. However, the maximum enrichment is still less than 4.5 wt%.

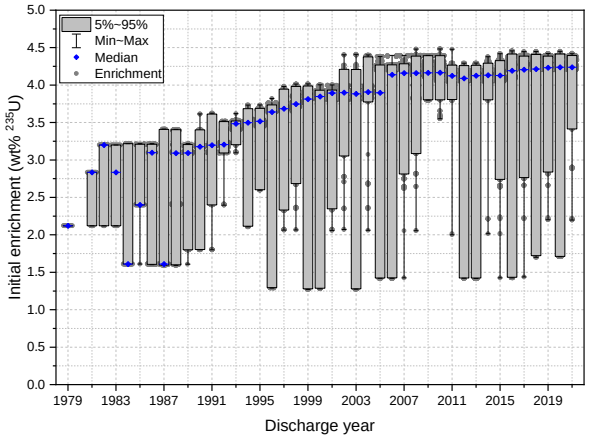

Fig. 3. Evolution of the initial enrichment as a function of fuel discharge year for SNFs

Fig. 4 shows the variation of median burnup and its ranges as a function of discharge year for SNFs. The discharge burnup of the fully burned fuel is correlated to the initial enrichment of fuel because the amount of fissile materials in an assembly limits the burnup that can be achieved. The second-generation fuels, ACE7 and PLUS7, were designed to have an allowable burnup of 55 MWd/kg and 55 MWd/kg or more, respectively [3]. Therefore, similar observations can be made for the evolution of burnup as for initial enrichment for both types of fuels. The median burnup generally increases over time, which is consistent with the trend of the median initial enrichment.

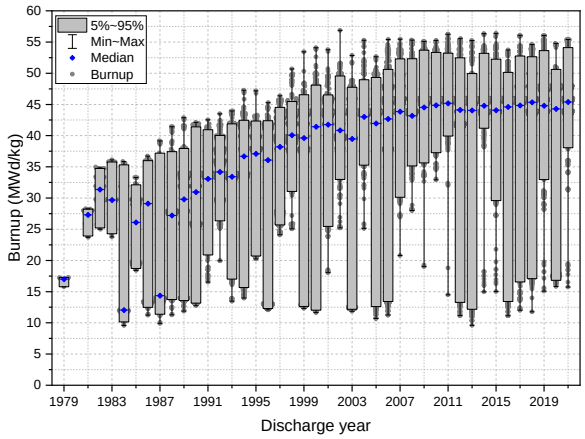

Fig. 4. Evolution of the median burnup as a function of fuel discharge year for SNFs

Fig. 5 shows the portion of the assembly types in the SNF database. There are four lattice types of domestic fuel assembly: WH 14×14, WH 16×16, WH 17×17, and CE 16×16. Of them, WH 17×17 and CE 16×16 type assemblies compose ~86 % of the total SNFs, and the portions of these type assemblies are nearly the same.

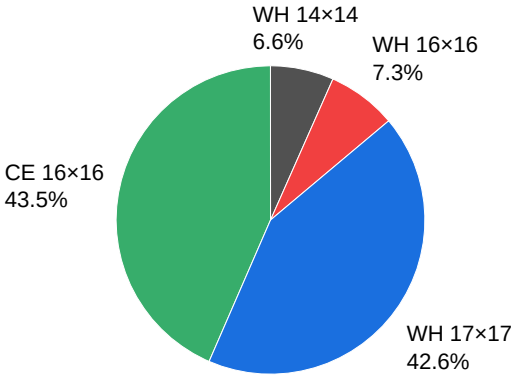

Fig. 5. Portions of assembly lattice types in SNF database

Fig. 6 shows the ranges of initial uranium inventory by fuel assembly lattice type. Among the WH 17×17 types, the maximum initial uranium loading is 468 kg, which is the highest among the total spent fuels in the database, while the maximum initial uranium inventory among the CE 16×16 type assemblies is 435 kg. The initial amount of uranium and burnup are important parameters for evaluating radioactivity, decay heat, criticality, radiation shielding safety, and inventory of SNFs.

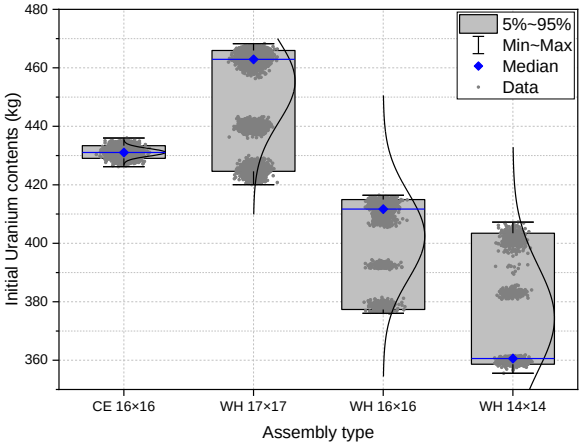

Fig. 6. Initial uranium content by assembly type

2.2 Design Basis Reference Spent Nuclear Fuel

A scenario was assumed in which 70 % of the total capacity of the deep geological disposal system was to be disposed of intermediate/low burnup SNFs, and the remaining 30 % of the SNFs with high burnup. As shown in fig. 2, when the SNFs are sorted based on the discharged burnup, the burnup corresponding to 70 % is 44.903 MWd/kg. Therefore, two design basis reference SNFs were selected based on discharge burnup. The first group (i.e., REF-45) was SNFs with a burnup less than 45 MWd/kg, and the reference burnup was set at 45 MWd/kg with conservatism. The second group (i.e., REF-55) was SNFs with a burnup of 45 MWd/kg or higher, and the reference burnup was set at 55 MWd/kg. The numbers of SNFs in each group are 14,826 and 6,144, respectively.

The initial enrichment and uranium inventory also should be determined for design basis reference SNF selection. Fig. 7 shows the enrichment distribution by the group. The median enrichment of the REF-45 group was 3.8 wt%, and the maximum value was 4.48 wt%. Enrichments were range from 4.0 wt% to 4.5 wt%, accounting for 34 % of this group. The median enrichment of the REF-55 group was 4.2 wt%, and the maximum value was 4.48 wt%, which is the same as that of the REF-45 group. Therefore, the reference enrichment of the REF-45 group and the REF-55 group was set to 4.5 wt% with conservatism.

Fig. 8 shows the uranium inventory distribution by the REF-45 and -55 groups. The total amounts of uranium in all SNFs in each group are 6,145 MTU and 2,719 MTU, respectively. The median values for the REF-45 and -55 groups are 431 kg and 432 kg, respectively, while the maximum value is the same as 468 kg. Therefore, the initial reference amount of uranium in the REF-45 group and the REF-55 group was set at 468 kg for conservatism.

In addition, it is necessary to set the specific power and effective full power day (EFPD) in the reactor core to calculate the source term of the SNFs. Assuming the specific power to be 40 MW/MTU, a typically used value, the EFPDs were calculated to satisfy the burnup of each group. Table I summarizes the characteristics of the determined design basis reference SNFs.

This study selected WH 17×17 ACE7 assembly as the design basis reference SNFs specification. However, of the 21 PWRs in operation in KOREA, six reactors are loaded with a WH 17×17 type, one reactor with a WH 16×16 type, and the remaining 14 reactors with a CE 16×16 type. Therefore, the proportion of CE 16×16 assemblies will increase, and accordingly, it is needed that the separated reference SNFs for WH 17×17 and CE 16×16 are considered and analyzed in future studies.

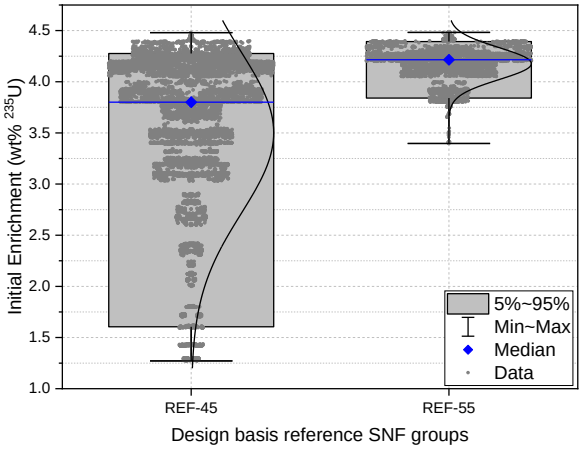

Fig. 7. Distribution of initial enrichment for design basis reference SNF groups

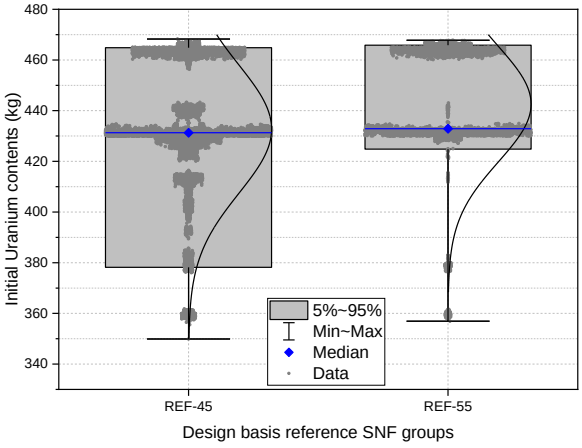

Fig. 8. Distribution of initial uranium inventory for design basis reference SNF groups

Table I: Design basis reference SNFs

Parameters		REF-45	REF-55
Burnup	[MWd/kg]	45	55
Initial enrichment	[wt% ^{235}U]	4.5	4.5
Initial uranium contents	[kg]	468	468
Specific power	[MW/MTU]	40	40
EFPDs	[days]	1125	1375

2.3 Modeling and Computational Method

SCALE 6.2.4 was used in this work to generate the reference values of the source terms. Specifically, the TRITON module in SCALE was used to generate the burnup-dependent effective one-group neutron cross section libraries, which were then used in ORIGEN standalone calculations [4].

In the burnup process, the neutron flux and neutron spectrum of the nuclear system affect the evolution of the nuclide compositions during irradiation. The TRITON sequence in SCALE takes into account these

interdependencies. For fuel irradiation, TRITON calls ORIGEN to perform the depletion and decay calculations after neutron flux calculation. TRITON allows users to save the burnup-dependent assembly-average neutron cross sections. These cross sections can be used in the standalone ORIGEN calculations. Multiple TRITON cases are usually run beforehand to cover the parameter space such as uranium enrichment and moderator density and to generate a set of ORIGEN cross section libraries. Customized cross section libraries are then generated by interpolating these pregenerated libraries using the specific parameter values for a particular ORIGEN case in ARP. The customized libraries were used in the ORIGEN calculations to generate the reference source term [5].

Fig. 9 illustrates the TRITON models for the WH 17×17 assembly configuration. Fuel rods, gaps, guide tubes, and cladding are modeled in detail.

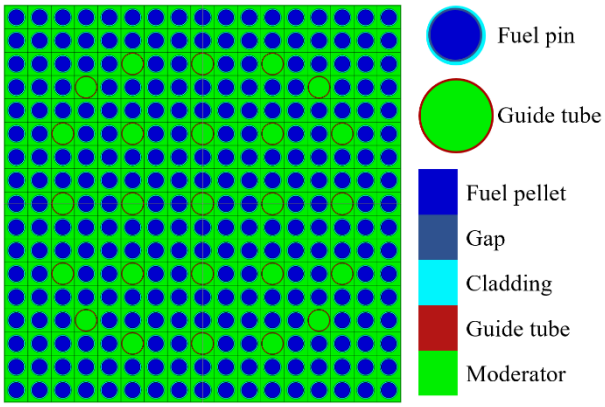

Fig. 9. Configuration of WH 17×17 assembly

2.4 Source Term Calculation Results

This section shows the results of source term parameters for the safety evaluation of the deep geological disposal system for the selected design basis reference SNFs. The results for the integral quantities are illustrated as a function of cooling time varying from the time of discharge to 1,000,000 years.

Fig. 10 shows the total activities for each group. The total activities of all groups decrease by three orders of magnitude in the first 250 years after discharge. At longer cooling times, the contribution to the activity from actinides (e.g., ^{241}Am, ^{241}Pu, ^{238}Pu, and ^{237}Np, etc.) and cesium isotopes (e.g., ^{137}Cs and ^{134}Cs) becomes more significant due to their longer half-lives. The isotopes are increasingly accumulated at higher burnups, so more considerable activities are observed in the fuels with higher burnups at long cooling times.

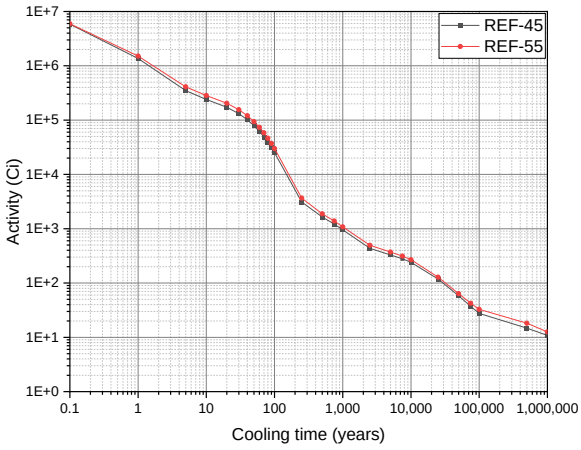

Fig. 10. Activity as a function of cooling time for each group

The results of total decay heat for each group are shown in Fig. 11. The decay heat decreases by two orders of magnitude 250 years after discharge. The results are similar at short cooling times (<0.1 years) among different burnups, but they begin to be different at longer cooling times (>1 year) because the decay heat is mainly driven by different sets of nuclides that have different half-lives. At longer cooling times, the relative contributions from actinides increase, and the fuels with higher burnups have higher actinide inventories [6]. Therefore, higher decay heats are observed at higher burnups, and the difference in decay heat between different burnup fuels is large, especially at longer cooling times.

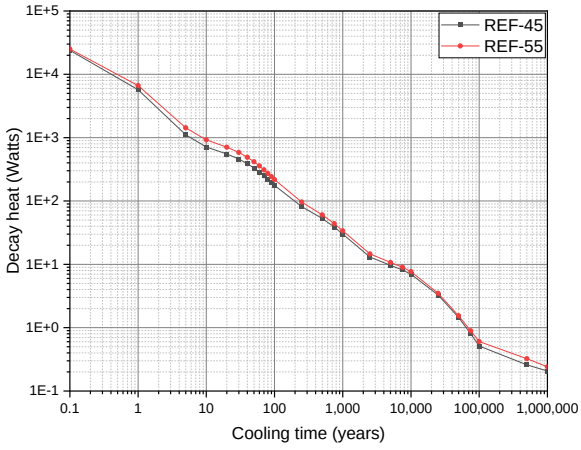

Fig. 11. Decay heat as a function of cooling time for each group

The gamma heat is the total energy of all emitted gamma rays from the decay of radionuclides in SNFs. This value is the gamma component of the decay heat power. The absorbed dose rate in air is nearly proportional to the energy of the emitted photon over the energy range of 70 keV to about 6 MeV. Therefore, the gamma heat is closely related to the unshielded dose rate near an SNF [4]. The results of gamma heat for each group are shown in Fig. 12. The gamma heat decreases

by four orders of magnitude at 250 years after discharge. After ~5 years of cooling time, the gamma radiation is mainly driven by the decay of ^{134}Cs (2-year half-life), ^{137}Cs (30-year half-life), and ^{154}Eu (8.6-year half-life), and the gamma radiation for each group increases with burnup. Therefore, the gamma heat from fuel with higher burnups is higher than that from low burnup fuels at cooling times longer than ~5 years.

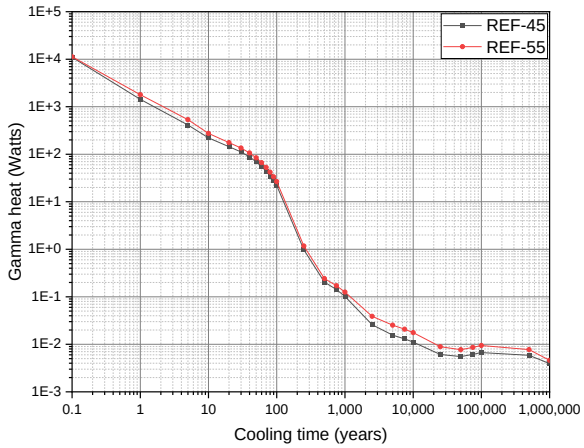

Fig. 12. Gamma energy release as a function of cooling time for each group

The results of total photon emission for each group are shown in Fig. 13. This is the total number of photons emitted in all energy groups considered. The photon emission decreases by nearly four orders of magnitude at 250 years after discharge. Short-lived fission products dominate the photon emission at short cooling times, then by ^{137}Cs, ^{154}Eu, and ^{134}Cs at intermediate cooling times and by mainly ^{137}Cs after ~5 years of cooling. Generally, the inventories of ^{137}Cs, ^{154}Eu, and ^{134}Cs increase with burnup, which leads to higher photon emissions in higher burnup fuels.

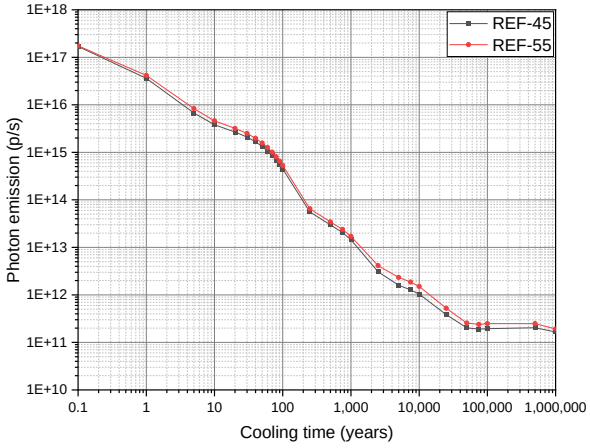

Fig. 13. Photon emission as a function of cooling time for each group

Fig. 14 shows the total plutonium inventory as cooling time after discharge. Because ^{239}Pu and ^{240}Pu, the two most abundant Pu isotopes, have long half-lives, the Pu total inventory decreases by only 10-20 % at 250 years after discharge. This decrease is mainly caused by the radioactive decay of ^{241}Pu (14-year half-life). While ^{238}Pu has a relatively short half-life (87.7 years), its abundance is relatively small. The amounts of all plutonium isotopes increase with burnup, except that ^{239}Pu has plateaued at high burnup.

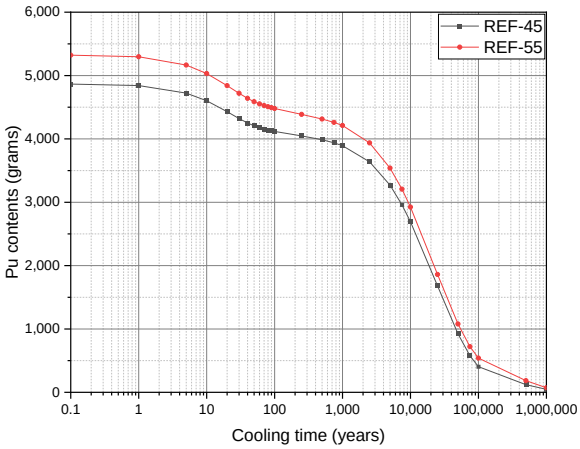

Fig. 14. Plutonium isotopes as a function of cooling time for each group

Fig. 15 and 16 show the 252-group neutron emission spectra and 47-group gamma spectra for the REF-45 group, respectively. It can be seen from the figures that the total neutron and gamma emission rate decreases as the cooling time increases. The spectrum can be used to determine the neutron and gamma fluxes on the SNFs surface, which are the parameters used in the nuclides release rate through the canister, or to calculate the surface neutron and gamma fluxes of a deep geological disposal canister which are used in radiation shielding analysis.

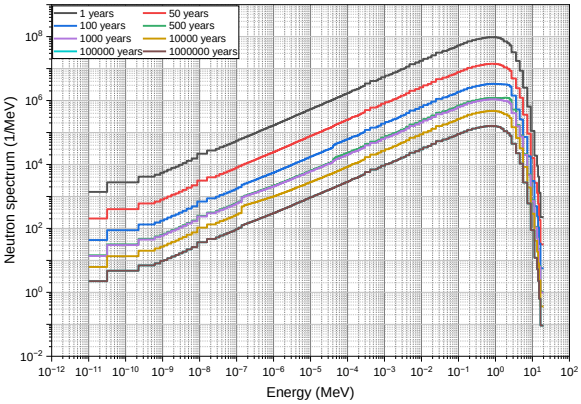

Fig. 15. 252-group neutron spectrum of REF-45 group

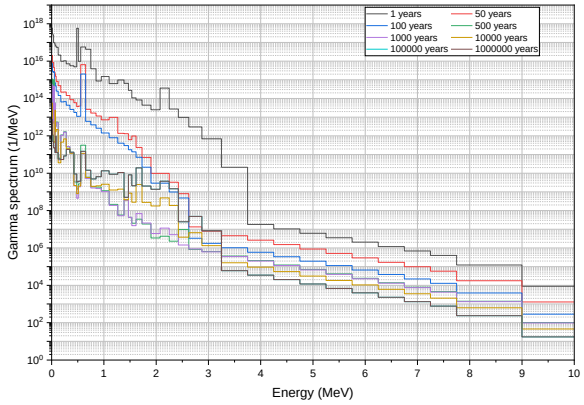

Fig. 16. 47-group gamma spectrum of REF-45 group

The decay heat emitted from actinides with a long half-life present in the SNFs and fission products with a shorter half-life than the actinides raise the temperature of the disposal canister, the buffer material, and the surrounding environment. It degrades the performance of the engineering and natural barriers in the disposal system. Therefore, when designing a disposal system, the decay heat must be kept to be sufficiently low in order to maintain the temperature below the temperature limit at which the performance of the engineered barrier is secured. In the leading countries for the deep geological disposal of SNFs, the canister decay heat limit of the KBS-3 disposal system in Sweden is 1,700 W [7], and the canister decay heat limit of the Olkiluoto site repository of POSIVA in Finland is regulated as 1,830 W [8]. The canisters of these two disposal systems were designed to dispose of four PWR assemblies. Since there have been no specific regulations and specific canister designs in our county, it is assumed that the four design basis reference SNFs determined in this study will be disposed of in the canisters of the previous two systems. The decay heat given in Fig. 11 is the value generated by a SNF with an initial uranium content of 468 kg. This value was normalized to the decay heat per 1 MTU, and it was assumed that four SNFs with the average value of initial uranium given in Fig. 8 are disposed of in a canister. Table II summarizes the average initial inventory of uranium for each group and the amount of uranium disposed of in the canister.

Fig. 17 shows the results of decay heat per canister as a function of cooling time for each group. The cooling time of interest was from 5 to 100 years after the SNFs were discharged from the reactor. As shown in Fig. 17, from the viewpoint of decay heat, it is expected that both the low/intermediate burnup group and the high burnup group can be disposed of in a disposal canister after 50 years of cooling time.

Table II: Summary of initial uranium contents

Group	Average U contents per SNF (gram)	Total U contents per canister (kg)
REF-45	432,720	1,730
REF-55	442,587	1,770

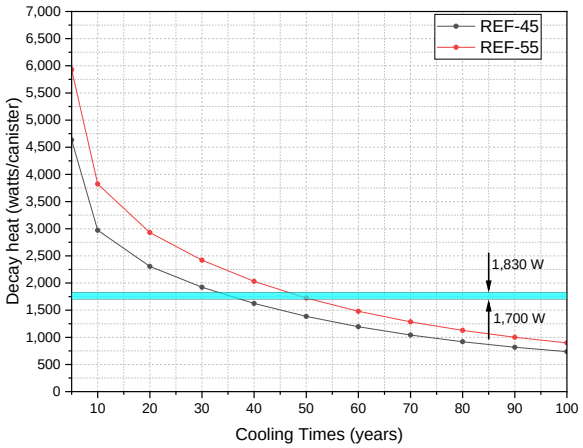

Fig. 17. Decay heat per canister as a function of cooling time for each group

3. Conclusions

In this work, the source terms were evaluated using the ORIGEN-ARP code to determine the design basis reference SNFs by analyzing the statistics of SNFs generated in Korea and deriving the input parameters for deep geological disposal system safety evaluation.

The number of domestic SNFs generated, initial enrichment, initial uranium inventory, discharge burnup, and irradiation history were used as the primary data for the generation of the source terms for the safety evaluation of the deep geological disposal system. It is assumed that 70 % of the capacity of the deep geological disposal system is loaded with low/intermediate burnup SNFs and the remaining 30 % with high burnup SNFs. As a result of statistical processing of the SNFs database by dividing it into two groups based on the burnup, which is the most important variable among the characteristics of SNFs, it was analyzed that 70 % of the SNFs generated so far have burnup of less than 45 MWd/kg. Therefore, based on this 45 MWd/kg burnup, the SNFs were divided into a low/intermediate burnup SNFs group and a high burnup SNFs group. Then, the design basis SNFs were determined by analyzing the initial enrichment, initial uranium content, and assembly type of the SNFs corresponding to each group. The intermediate/low burnup group was 45 MWd/kg burnup, 4.5 wt% initial enrichment, and 468 kg initial uranium inventory, while the high burnup group was 55 MWd/kg, 4.5 wt% initial enrichment, and 468 kg initial uranium inventory.

For each design basis SNFs, the ORIGEN-ARP code was used to evaluate the decay heat, which is essential for the safety evaluation of the deep geological disposal system, and the neutron and gamma emission spectra required for radiation shielding analysis. And a database on the results of these quantities for source term is produced. Also, it was shown that from the viewpoint of decay heat, both the low/intermediate burnup group and the high burnup group could be disposed of in a disposal canister after 50 years of cooling time.

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