Evaluation of Radionuclide inventories and Instant Release Fraction of Low-Enriched PLUS7 Type Fuel Assembly

Kwang Pyo Choi [a], and Ser Gi Hong [a,*]
[a] *Department of Nuclear Engineering, Hanyang University*
*Author: sskk1216@hanyang.ac.kr, *Corresponding author: sergihong@hanyang.ac.kr*

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1. Introduction

In South Korea, spent nuclear fuels from PWR are currently stored in on-site wet storage facilities. Since 2030, several wet spent fuel storages will be saturated, which are started from Hanbit (2030) followed by Hanul (2031), Kori (2032), and Shin Wolseong (2042). [1] This impending saturation has prompted consideration of various final disposal methods, with safety assessment being the most critical thing to be considered. Especially many countries have considered Deep Geological Repository (DGR) concept. Within the safety assessment framework, the source term evaluation stands as the most crucial component. The source term for disposal facilities involve two key processes: 1) Evaluating the inventory of radionuclides within spent nuclear fuel that significantly impact the safety assessment and 2) Assessing how these radionuclides might be released in the event of canister failure.

To select what nuclides' inventory to be evaluated, we've referenced the several reports and papers about selecting of safety-relevant-radionuclides. [1] In general, it is considered that there're two main mechanisms for radionuclide release, which are the congruent release and the instant release. The congruent release represents a slow process where radionuclides uniformly distributed within the fuel matrix are released as the fuel dissolves in water, catalyzed by the products like hydrogen peroxide produced by radiolysis, while the instant release represents a rapid process where radionuclides partially distributed in the spent nuclear fuel are immediately released due to significant defects in the canister. [2]

The main point of this paper is to evaluate the safety-relevant radionuclide inventory of the reference spent nuclear fuel, specifically low-burnup PLUS7 type fuel, expected to be discharged at the time of fuel storage saturation in the aspect of DGR concept. Furthermore, it extends to generating Instant Release Fraction(IRF) values for some radionuclides, including ^{14}C, ^{59}Ni, ^{63}Ni, ^{93}Zr, ^{93}Mo, ^{93m}Nb, and ^{94}Nb.

2. Methodologies

2.1. Reference PLUS7 low-burnup fuel assembly

This work considered 56 representative radionuclides which were selected by our group to generate the source terms for the reference PWR SNFs [1]. The previous work selected four reference PWR SNFs with considering high and low burnup for CE16x16 and WH 17x17 types. As a preliminary work, this work focused only on the low-burnup PLUS7 type reference SNF within the CE 16×16 category, which accounts for ~43.5% of the PWR SNFs in Korea. The parameters of the reference SNF are specified in Table I.

Table I. Parameters of the reference PLUS7 fuel assembly

Parameter	REF_PLUS7_LB
Lattice type	16×16
Burnup (MWd·kg^{-1})	45
Initial uranium enrichment (%)	4.5
Initial uranium loading (kg)	436
Specific power (MW·MTU^{-1})	40
Mid Grid	
Material	ZIRLO
Numbers per assembly	9
Weight each, kg	0.852
Top, Bottom Grid	
Material	Inconel 718
Numbers per assembly	2 (top, bottom)
Weight each, kg	0.651
Protective Grid	
Material	Inconel 718
Numbers per assembly	1
Weight each, kg	0.415
Bottom nozzle	
Material	SS 304
Numbers per assembly	1
Weight each, kg	5.4
Top nozzle	
Material	SS 304 + Inconel 718
Numbers per assembly	1
Weight each, kg	16.8

The most data of structural materials are referred from the publicly available Final Safety Analysis Report(FSAR) of Hanbit unit 3, 4 [3]. We only consider spacer grids, protective grids, top and bottom nozzles as the structural material. The mass data of top and bottom nozzle are referred from the ORNL's reference information for SNF radiation source term calculations [4]. In particular, for the top nozzle, we homogenized

SS 304 parts and Inconel 718 parts due to the lack of geometrical information. The configurations are specified in **Fig. 1** and **Fig. 2** for the unit fuel cell and assembly including axial and radial cut views, respectively.

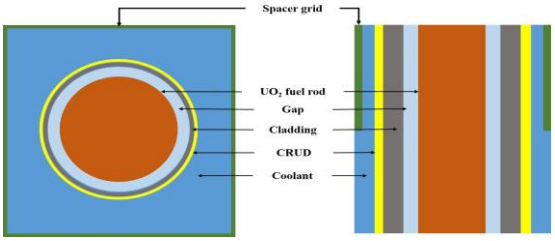
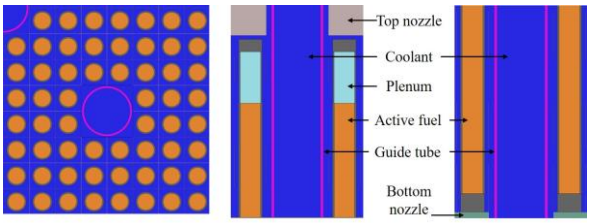


Fig. 1. Radial and axial configuration of PLUS7 fuel rod



<Radial> **<Upper end>** **<Lower end>**

Fig. 2. Radial and axial configuration of PLUS7 fuel assembly

The simplified radial configuration of the top nozzle and bottom nozzles is specified in **Fig. 3.**

Fig. 3. Radial configuration of Bottom and Top nozzle lattice

Also, we model Chalk River Unidentified Deposits (CRUD) geometry on the outer surface of 236 fuel rods. Because of the turbulence of the high temperature coolant nearby cladding, CRUD deposits on cladding surface within upper ~ middle axial position of active fuel length. And we assume the uniform 7.5 μm thickness on the whole active fuel length (381cm) to calculate conservative CRUD inventory [5]. Detail information of CRUD composition is in **Table II**.

Table II. Composition of CRUD

Component	weight Fraction [%]
Ni	25.01
Fe	44.67
O	28.66
Cr	0.86
Mn	0.8

Additionally, we considered the impurity concentrations in UO_2, ZIRLO, and Inconel 718 and SS 304 to realistically calculate radionuclide inventories for N, Cl, [6] and Ni. [7] The impurity concentrations are listed in **Table III**.

Table III. Impurity composition in materials

Elements	Weight fraction [ppm]		
	UO_2	ZIRLO	Inconel 718, SS304
N	75	80	1000
Cl	25	10	10
Ni	400	80	80

2.2. Partitioning component of source term and safety-relevant radionuclides

To evaluate source term of radionuclide transport, it is needed to partition the fuel assembly into four parts: fuel matrix, cladding, structural parts, and CRUD. The fuel matrix includes fission products, actinides, and activation products from UO_2 fission, including some volatile elements diffused into the fuel-cladding gap and UO_2 grain boundaries. Cladding refers to the activation products in the cladding. Structural parts includes the activation products in the guide tubes, spacer grids, top nozzle, and bottom nozzle. CRUD includes activation products in the CRUD deposition on the cladding surface.

We focus on 56 safety-relevant radionuclides based on specific screening process. Radionuclides lists are in **Table IV**. [1]

Table IV. List of the safety-relevant radionuclides

Fission and Activation products					
^{108m}Ag	^{14}C	^{113m}Cd	^{36}Cl	^{134}Cs	^{135}Cs
^{137}Cs	^{152}Eu	^{154}Eu	^{155}Eu	^{3}H	^{129}I
^{93}Mo	^{93m}Nb	^{94}Nb	^{59}Ni	^{63}Ni	^{107}Pd
^{147}Pm	^{125}Sb	^{79}Se	^{151}Sm	^{121m}Sn	^{126}Sn
^{90}Sr	^{99}Tc	^{93}Zr			
Radionuclides from decay chain					
4N					
^{244}Cm	^{240}Pu	^{236}U	^{232}Th	^{232}U	^{228}Th
4N+1					
^{249}Cf	^{245}Cm	^{241}Pu	^{241}Am	^{237}Np	^{233}U
^{229}Th					
4N+2					
^{250}Cf	^{246}Cm	^{242}Pu	^{242m}Am	^{242}Pu	^{238}U
^{238}Pu	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	
4N+3					
^{243}Am	^{243}Cm	^{239}Pu	^{235}U	^{231}Pa	^{227}Ac

The 20 radionuclides which are instantly released are listed **table V**. [9]

Table V. Instantly Released Radionuclides Groups

Group	Radionuclides
Group 1 (Very volatile radionuclide)	^{129}I, ^{135}Cs, ^{137}Cs, ^{36}Cl, ^{79}Se
Group 2 (Volatile radionuclide)	^{107}Pd, ^{126}Sn, ^{90}Sr, ^{99}Tc
Group 3 (Instantly released only from CRUD)	^{93}Mo, ^{93m}Nb, ^{94}Nb, ^{59}Ni, ^{63}Ni, ^{93}Zr
Group 4 (Individually treated)	^{108m}Ag, ^{3}H, ^{14}C, ^{113m}Cd, ^{121m}Sn

For Group 1, we already evaluated IRF values in last year. Also, due to the lack of leaching data of group 2 and ^{121m}Sn, we decided to apply the SKB's data. And, for ^{108m}Ag and ^{113m}Cd, we don't evaluate IRF value, because it is not considered to dispose the control rod together in the canister in the Korean deep geological concept. [9]

So, we calculate the IRF values of ^{14}C, ^{59}Ni, ^{63}Ni, ^{93}Zr, ^{93}Mo, ^{93m}Nb, and ^{94}Nb in this work. For ^{14}C, it is generated considered that 10% of fuel Matrix and 20% of Cladding inventory are instantly released. On the other hand, for ^{59}Ni, ^{63}Ni, ^{93}Zr, ^{93}Mo, ^{93m}Nb, and ^{94}Nb, only CRUD inventories are considered to be instantly released. The calculation methods of IRFs of each nuclide are listed in **Table VI**. [8]

Table VI. Some Radionuclides' calculation method of IRF

Radionuclides	IRF formula
^{14}C	$(0.1\times I_{UO2} + 0.2\times I_{Cladding}) / I_{tot}$
^{59}Ni, ^{63}Ni, ^{93}Zr, ^{93}Mo, ^{93m}Nb, ^{94}Nb	I_{CRUD} / I_{tot}

I_{tot} is the total inventory, I_{UO2} is Fuel matrix inventory, $I_{Cladding}$ is Cladding inventory, and I_{CRUD} is CRUD inventory of a radionuclide. All inventories' unit is [Bq].

2.3. Computational method

The fuel assembly burnup calculations were performed using SERPENT2, a famous Monte Carlo neutron transport and burnup calculation code developed by VTT. [10] For neutron transport calculations, we utilized the ENDF/B-VII.1 point-wise cross section library. In the 3-D fuel assembly calculations, the top nozzle was homogenized while the remaining parts were treated heterogeneously. The CRAM (Chebyshev Rational Approximation Method) option was applied for burnup depletion calculation. A burnup time step of 45 days implemented, with two 30-day downtime periods applied after 375 EFPDs and 750 EFPDs. [1] Additionally, a 50-year cooling time was incorporated to reflect the decay heat condition of disposal of SNF. The simulation parameters included 500 active cycles and 100 inactive cycles, with 75000 histories applied to each cycle.

3. Results

3.1. Radionuclide inventories

The results of the evaluated inventories for the considered radionuclides are listed in the **table VII**.

Table VII. Calculation results of radionuclide inventories in the PWR SNF Canister

Radio-nuclide	Activity [Bq/canister]				
	Total	Fuel Matrix	Cladding	Structural parts	CRUD
^{3}H	8.83E+09	7.11E+09	1.41E+09	3.05E+08	4.27E+05
^{14}C	1.58E+11	1.26E+11	2.34E+10	8.39E+09	1.19E+08
^{36}Cl	4.26E+09	3.84E+09	3.33E+08	8.42E+07	1.70E-18
^{59}Ni	5.34E+10	1.20E+10	5.19E+08	2.29E+10	1.80E+10
^{63}Ni	5.65E+12	1.35E+12	5.87E+10	2.31E+12	1.93E+12
^{79}Se	6.29E+09	6.29E+09	1.68E-11	3.43E-12	0
^{90}Sr	1.97E+15	1.97E+15	4.32E+08	8.08E+07	0
^{93}Zr	1.67E+11	1.57E+11	8.31E+09	1.73E+09	0
^{93}Mo	2.01E+08	5.83E+04	2.43E+04	2.01E+08	0
^{93m}Nb	1.45E+11	1.36E+11	7.22E+09	1.66E+09	0
^{94}Nb	4.05E+11	2.22E+07	3.34E+11	7.08E+10	0
^{99}Tc	1.16E+12	1.16E+12	9.46E+05	2.70E+07	0
^{107}Pd	9.21E+09	9.21E+09	3.21E-06	6.17E-07	0
^{108m}Ag	7.71E+05	7.71E+05	5.99E-04	1.23E-04	0
^{113m}Cd	1.84E+11	1.84E+11	3.80E+04	7.88E+03	0
^{121m}Sn	1.21E+12	1.18E+12	3.14E+10	6.13E+09	0
^{126}Sn	2.32E+10	2.32E+10	3.98E+03	8.03E+02	0
^{129}I	2.35E+09	2.35E+09	0	0	0
^{135}Cs	3.97E+10	3.97E+10	0	0	0
^{137}Cs	2.88E+15	2.88E+15	0	0	0
^{134}Cs	8.03E+08	8.03E+08	0	0	0
^{152}Eu	1.66E+10	1.66E+10	0	0	0
^{154}Eu	1.11E+13	1.11E+13	0	0	0
^{155}Eu	2.34E+11	2.34E+11	0	0	0
^{147}Pm	2.58E+10	2.58E+10	0	0	0
^{125}Sb	3.26E+09	3.05E+09	1.77E+08	3.42E+07	0
^{151}Sm	1.67E+13	1.67E+13	0	0	0
^{244}Cm	4.30E+13	4.30E+13	0	0	0
^{240}Pu	3.87E+13	3.87E+13	0	0	0
^{236}U	2.51E+10	2.51E+10	0	0	0
^{232}Th	6.42E+01	6.42E+01	0	0	0
^{232}U	3.54E+08	3.54E+08	0	0	0
^{228}Th	3.64E+08	3.64E+08	0	0	0
^{249}Cf	1.63E+06	1.63E+06	0	0	0
^{245}Cm	4.34E+10	4.34E+10	0	0	0
^{241}Pu	9.82E+14	9.82E+14	0	0	0
^{241}Am	3.26E+14	3.26E+14	0	0	0
^{237}Np	3.33E+10	3.33E+10	0	0	0
^{233}U	9.68E+06	9.68E+06	0	0	0
^{229}Th	3.90E+04	3.90E+04	0	0	0
^{250}Cf	1.75E-02	1.75E-02	0	0	0
^{246}Cm	6.52E+09	6.52E+09	0	0	0
^{242m}Am	2.96E+12	2.96E+12	0	0	0
^{242}Pu	1.68E+11	1.68E+11	0	0	0
^{238}U	2.00E+10	2.00E+10	0	0	0
^{238}Pu	2.20E+14	2.20E+14	0	0	0

Table VII. Calculation results of radionuclide inventories in the PWR SNF canister (continued)

Radio-nuclide	Activity [Bq/canister]				
	Total	Fuel Matrix	Cladding	Structural parts	CRUD
^{234}U	1.24E+11	1.24E+11	0	0	0
^{230}Th	5.11E+07	5.11E+07	0	0	0
^{226}Ra	5.43E+05	5.43E+05	0	0	0
^{210}Pb	2.03E+05	2.03E+05	0	0	0
^{243}Am	1.75E+12	1.75E+12	0	0	0
^{243}Cm	6.37E+11	6.37E+11	0	0	0
^{239}Pu	2.50E+13	2.50E+13	0	0	0
^{235}U	1.56E+09	1.56E+09	0	0	0
^{231}Pa	3.70E+06	3.70E+06	0	0	0
^{227}Ac	2.48E+06	2.48E+06	0	0	0

This calculation can determine which nuclides are abundant in which part of PLUS7 fuel assembly, and can be used to calculate the accurate release amount of radionuclides by applying the different release rate of each part in the DGR situation. So we calculate radionuclide inventories of 4 assemblies in the PWR SNF canister like SKB and POSIVA. [6], [11]

3.2. IRF calculation based on the radionuclide inventories of PLUS7 fuel assembly

Based on **table VII**, we calculate the IRF of the radionuclides in **table VI**. Calculated IRFs are listed in **table VIII**.

Table VIII. Calculation results of IRF

Radionuclides	IRF [%]
^{14}C	10.94
^{59}Ni	33.58
^{63}Ni	34.07
^{93}Zr	0
^{93}Mo	0
^{93m}Nb	0
^{94}Nb	0

For ^{14}C, the calculation result is bit higher than the range of SKB's data which is 5.0% as the best estimate and 10.0% as the bound estimate. [8]

For ^{59}Ni and ^{63}Ni, the calculation result shows a significant discrepancy with SKB's data which is 0.16% and 0.15% for each. This discrepancy can be explained with several things. [8] First, It is attributed to our assumption that CRUD formation occurred along the entire active fuel length using the maximum CRUD thickness proposed in the referenced paper. [5] Second, in SKB's data, control rod's inventory was included in IRF calculation, and there're more lot of Ni inventories in the structural material than our calculation. [11]

For ^{93}Zr, ^{93}Mo, ^{93m}Nb, and ^{94}Nb, our results show 0% for all IRFs of them. But SKB's data shows less than 0.0001% for all IRFs of them. For the results except ^{59}Ni and ^{63}Ni, it could be difference in the CRUD composition of SKB and our assumption which is the Ni-Fe oriented composition of CRUD. [8]

4. Conclusion and future works

In this work, we calculated the radionuclide inventory and the IRF for selected nuclides in low-burnup PLUS7 fuel assembly, assuming DGR concept of the disposal of SNF in Korea. To produce more accurate IRF and inventory data, it is evident that more precise specifications for both the fuel assembly and CRUD are primarily required. Furthermore, once accurate specifications are established, cross-validation using alternative computational methods will be necessary to ensure the reliability of our results.

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