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

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Contents

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❑ Necessity of Safety Analysis of Deep Geological Repository

- Currently, the PWR Spent Nuclear Fuel(SNF)s in Korea are stored in the wet storages at the individual reactor site.
- Decision of the final repository concept is postponed, and the wet storages are going to be saturated soon.[1)]
- In 2030, the initial operation license of 10 Nuclear Power Plants is going to be expired, and the current government makes the possibility of continued operation of those be opened.[2)]
- The interim storage is the method of storing the SNFs with a clear limit.
- Many countries consider a Deep Geological Repository(DGR) as a final repository concept.
- At first, we need to preliminarily analyze the source term of the DGR in Korea.

Figure 1. Nuclear Power Plant(NPP) Operation Status in Korea (24.04.05)[2)]

Table 1. Estimated saturation point by nuclear power plant site (unit: year)[1)]

	KORI (PWR)	HANBIT (PWR)	HANUL (PWR)	SAEUL (PWR)	SHIN-WOLSONG (PWR)	WOLSONG (CANDU)
Basic Plan ('21.12)	2031	2031	2032	2066	2044	-
Renewed ('23.2)	2032*	2030	2031	2066	2042	2037

* KORI-2 SFP forecasted to saturate by 2028 without high-density rack installation

1) 산업통상자원부, "사용후핵연료 발생량· 포화전망 설명회", 2023년2월10일
2) 한국원자력학회, 원자력 소식, 한국 원전의 계속운전 현황, 2024년4월5일

1. INTRODUCTION (2/3)

❑ Source Term Analysis

- In the DGR concept of SKB and POSIVA, 4 PWR SNFs are stored in the single copper canister.[3)]
- The source term is in the SNFs and construction materials.
- The source term analysis includes the calculation below:
  - Radionuclide inventory, Radioactivity
  - Decay heat
  - Neutron/Gamma spectrum
- Radionuclide release amount and release rate can be calculated through the source term analysis.

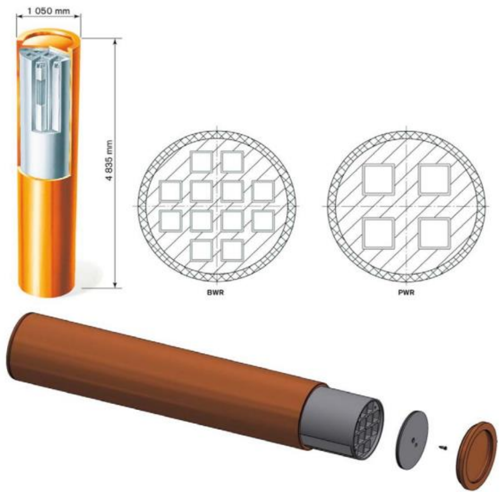


Figure 2. Schematic of the general design of the KBS-3 canister[3)]

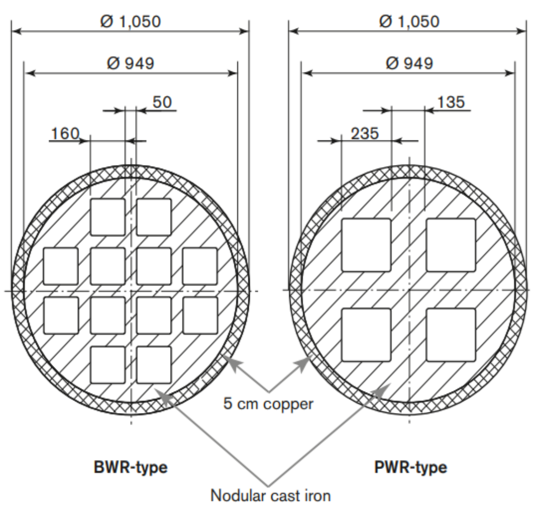
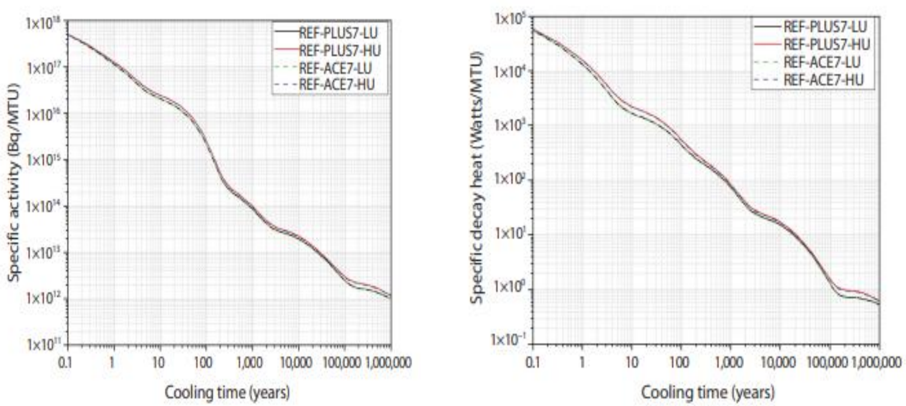


Figure 3. SKB's reference canister[4)]



Radioactivity (Bq/MTU)

Decay Heat (Watts/MTU)

Figure 4. Examples of the results of the source term analysis[5)]

3) Mikael Jonsson et al., Mechanical design analysis for the canister, POSIVA SKB Report 04, 2018
4) Johan Andersson et al., Data report for the safety assessment SR-site, SKB TR-10-52, Sweden, 2010
5) K. J. Choi, S. S. Oh, and S. G. Hong, "Preliminary Selection of Safety-Relevant Radionuclides for Long-term Safety Assessment of Deep Geological Disposal of Spent Nuclear Fuel in South Korea", JNFCWT, Vol.21, No.4, p.451-463, 2023

❑ Radionuclide Release Mechanism

- There are two major mechanisms:[4,6)]
  - **Instant Release :** Release process of partially distributed nuclides from a fuel-cladding gap, fuel grain boundary, CRUD, and Cladding etc. (Rapid)
  - Congruent Release : Release process from fuel matrix due to the dissolution of fuel contacting underwater. (Very slow)
- Especially, the fraction of the instantly released amount to the total amount is the **Instant Release Fraction (IRF).**
- **In this work, those two were calculated :**
  - **IRF**
  - **Radionuclide inventory :** the inventory of safety-relevant radionuclides in the single canister
- The Serpent 2 Monte-Carlo depletion calculation code is used.

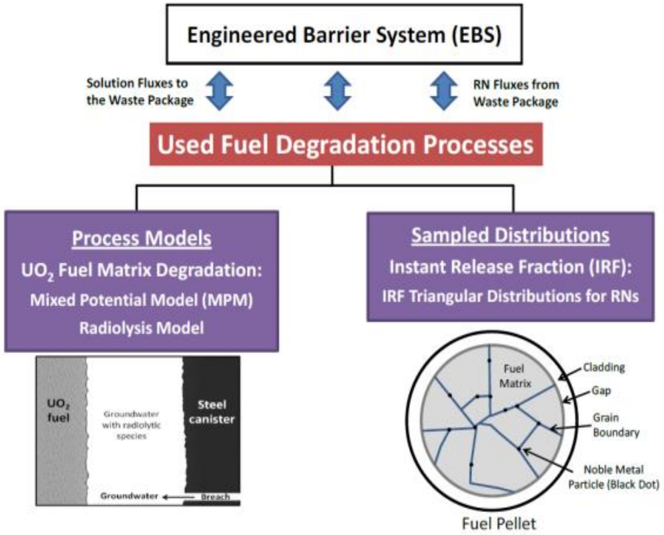
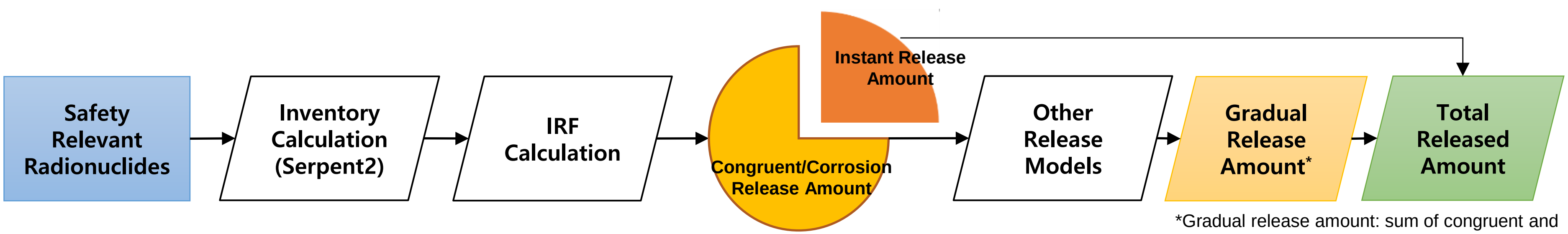


Figure 5. Radionuclides release model in EBS[7)]



*Gradual release amount: sum of congruent and corrosion release amounts at a certain time

Figure 6. Schematic figure of application IRF in radionuclide release model

6) Posiva Oy, Safety Case for the Disposal of Spent Nuclear Fuel at Olkiluoto – Models and Data for the Repository System 2012, Posiva 2013-01, 2013
7) Sandia National Laboratories(2012), Integration of EBS Models with Generic Disposal System Models, SAND2012-7762 P

❑ Instant Release Radionuclides

- 18 radionuclides are considered as an instant release radionuclide.
- Instant release radionuclides are distributed to 4 groups[4].
- In previous work, Group 1 radionuclides' data was evaluated.[8]
- In this work, we calculated only the **IRF of ^{14}C, ^{59}Ni, ^{63}Ni, ^{93}Zr, ^{93}Mo, ^{93m}Nb, ^{94}Nb.**
- For ^{14}C, 10% of UO_2 inventory and 20% of the cladding inventory are instantly released.
- For ^{59}Ni, ^{63}Ni, ^{93}Zr, ^{93}Mo, ^{93m}Nb, ^{94}Nb, only CRUD inventory is instantly released.

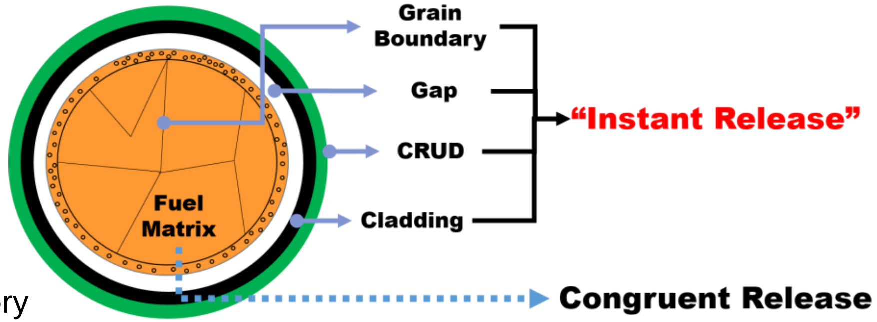


Figure 7. locations of the instant release in spent fuel pellet

Table 2. Instant Release Radionuclides' group[4]

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)	^{3}H, ^{14}C, ^{121m}Sn

Table 3. IRF formula of some radionuclides[4]

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

I_{tot} : Total Radioactivity
I_{UO2} : Radioactivity in UO2 matrix
I_{clad} : Radioactivity in Cladding
I_{crud} : Radioactivity in CRUD

8) K. P. Choi, and S. G. Hong, A Preliminary Study for Evaluation of Instant Release Fractions for PWR Spent Fuels Based on Literature Review, *Korean Radioacive Waste Society Autumn Conference*, 2023

2. METHODOLOGY (2/4)

Safety-relevant radionuclide

- In previous work, 56 safety-relevant radionuclides were selected.[5)]
- Those radionuclides are distributed in the each part of the canister.
- There're 4 components in the single canister
  - UO_2, Cladding, CRUD, Construction materials
- It is necessary to calculate each radioactivity of the components.[4,6)]

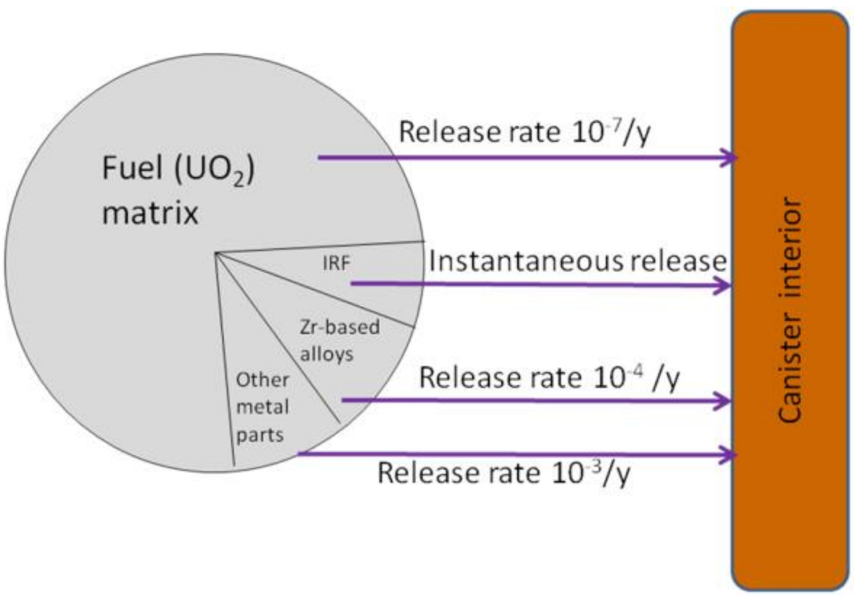


Figure 8. Chart of the source term as inventory components[6)]

Table 4. 56 Safety-relevant radionuclides[5)]

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					
^{244}Cm	^{240}Pu	^{236}U	^{232}Th	^{232}U	^{228}Th
^{249}Cf	^{245}Cm	^{241}Pu	^{241}Am	^{237}Np	^{233}U
^{229}Th	^{250}Cf	^{246}Cm	^{242}Pu	^{242m}Am	^{238}U
^{238}Pu	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{243}Am
^{243}Cm	^{239}Pu	^{235}U	^{231}Pa	^{227}Ac	

❑ Serpent2 modeling of Reference PLUS7 low-burnup fuel assembly

- In previous work, the 4 type of reference PWR SNFs are selected.[5)]
- Preliminary, Reference low-burnup PLUS7 assembly's radioactivity is calculated.
- Construction materials:
  - Guide tubes, Instrumental tube, Spacer grids, Nozzles
- Parameters of the FSAR of Hanbit unit 3, 4 are used.[8)]
- For some parameters, Other reference data is used.[9)]

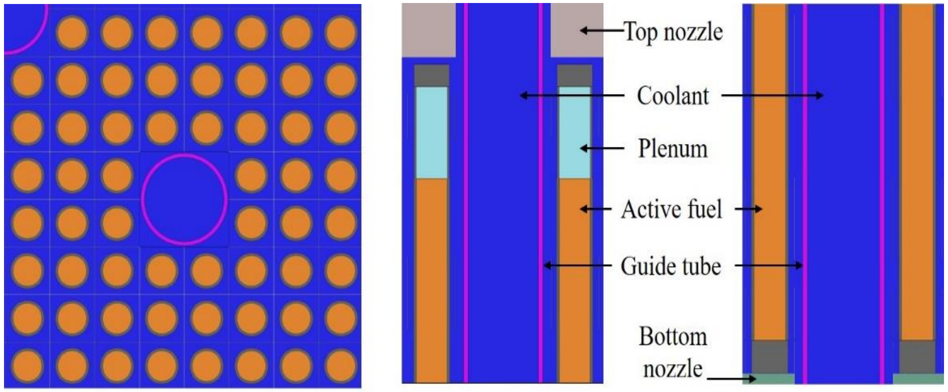


Figure 9. Configuration of PLUS7 fuel assembly

Table 5. Parameters of reference Low-Burnup PLUS7 assembly[9,10)]

Parameter		REF_PLUS7_LB
Lattice type		16×16
Burnup (MWd·kg^{-1})		45
Initial uranium enrichment (wt.%)		4.5
Initial uranium loading (kg)		436
Specific power (MW·MTU^{-1})		40
Mid Grid	Material	ZIRLO
	Number	9
	Weight each, kg	0.852
Top, Bottom Grid	Material	Inconel 718
	Number	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

Table 6. N, Cl, Ni Impurity composition[11, 12)]

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

9) 한국수력원자력㈜, 한빛 3,4호기 최종안전성 분석보고서 공개본, 4장
10) Georgeta Radulescu, Brandon R. Grogan, and Kaushik Banerjee, Fuel Assembly Reference Information for SNF Radiation Source Term Calculations, ORNL/SPR-2021/2093, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 2021
11) Posiva Oy, Source Terms for the Safety Case in Support of the Operating Licence Application, POSIVA 2021-11
12) Häkkinen, Silja, Impurities in LWR fuel and structural materials, VTT-R-00184-20

2. METHODOLOGY (4/4)

Serpent2 modeling of Reference PLUS7 low-burnup fuel assembly

- The Chalk River Undefined Deposits(CRUD) on the cladding surface is also considered
- Most of the CRUD is deposited on the upper part of active fuel length[13)]
- So uniform thickness is assumed as 7.5 μm, the half of the maximum thickness[14)]
- Averaged fraction is applied to the composition of the CRUD

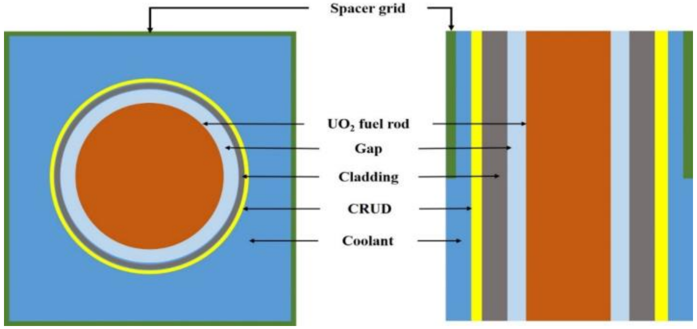


Figure 12. CRUD geometry in Serpent2

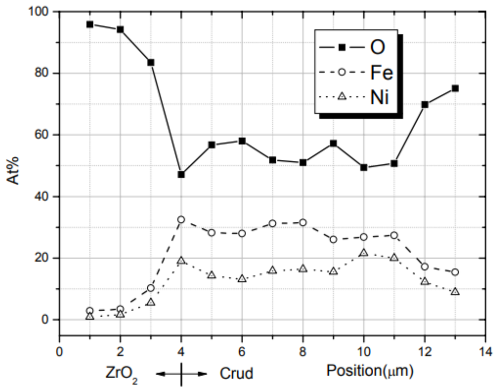


Figure 10. CRUD chemical composition distribution at each position[11)]

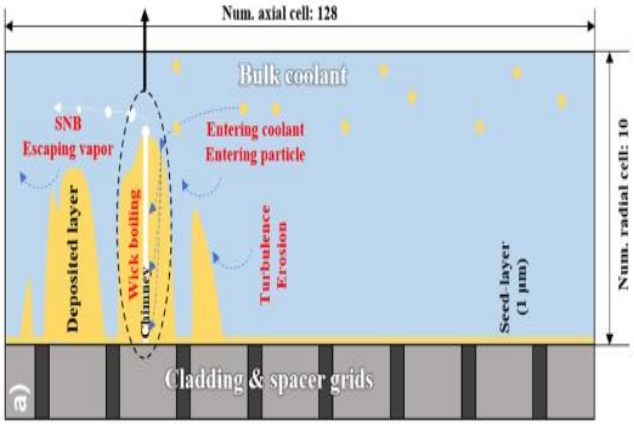


Figure 11. Vertical CRUD geometry on Cladding[10)]

Table 7. Averaged CRUD element composition

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

13) S. J. Seo, N. K. Chae, Samuel Park, Richard I. Foster, S. Y. Choi, "Modeling of deposition and erosion of CRUD on fuel surfaces under sub-cooled nucleate boiling in PWR"
14) H. M. Kwon, H. S. Seo, Y. H. Jung, D. K. Min, and Y. B. Chun, "Study of the Composition of Cruds on the High Burnup Fuel Cladding in PWR"

3. RESULTS (1/3)

Radioactivity and IRF value

- Radioactivity and IRF of the radionuclides which have a IRF value

Table 8. Radioactivity of nuclides which have IRF value

Radio-nuclide	Radioactivity [Bq/canister]				
	Total	Fuel Matrix	Cladding	Stuructural Materials	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

Table 9. Calculation results of IRFs

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

Radioactivity

- Radioactivity of the radionuclides which have zero IRF value

Table 10. Radioactivity of nuclides(Non-IRF)

Radio-nuclide	Radioactivity [Bq/canister]				
	Total	Fuel Matrix	Cladding	Stuructural Materials	CRUD
^{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

3. RESULTS (3/3)

❑ Radioactivity

Table 10. Radioactivity of nuclides(Non-IRF, continued)

Radio-nuclide	Radioactivity [Bq/canister]				
	Total	Fuel Matrix	Cladding	Stuructural Materials	CRUD
^{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
^{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

4. CONCLUSIONS (1/1)

Necessity of the data verification for the Safety analysis

- In this study, we calculated the radionuclide activities and instant release fractions (IRFs) for selected nuclides in a reference low-burnup PLUS7-type fuel assembly.
- In comparison with the SKB's IRF data[4)] :
  - For ^{14}C, it was similar level
  - For, ^{59}Ni, ^{63}Ni, it was very overestimated
  - For ^{93}Mo, ^{93m}Nb, ^{94}Nb, ^{93}Zr, SKB's data is also small, but our data is literally zero.
- From these results, more research about the modeling the CRUD in computational code is needed
- Also, the source term analysis has a main role of guarantying the precision of the Safety analysis
- For this sake, Cross-validation is needed between this result and other result from using another computational code like SCALE, DeCART2D

Table 11. Comparison between our results and SKB's data[4)]

Radionuclides	IRF [%]	IRF [%] (SKB)
^{14}C	10.94	11
^{59}Ni	33.58	0.16
^{63}Ni	34.07	0.15
^{93}Zr	0	7.0×10^{-6}
^{93}Mo	0	5.5×10^{-3}
^{93m}Nb	0	6.9×10^{-5}
^{94}Nb	0	6.8×10^{-5}

Thank you for your attention.