Possibility of Disposal for Spent Nuclear Fuel Reprocessing in the Aspect of the Radiological Risk of Human Intrusion

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1. Introduction

Scenarios after the closure of disposal facilities are classified mainly into situations due to natural phenomena and human intrusion by the International Commission on Radiological Protection (ICRP). Scenarios by natural phenomena are divided into normal scenarios that occur when the components of the disposal facility perform safety functions as designed and abnormal scenarios caused by natural phenomena such as earthquakes and floods. Scenario by human intrusion is caused by actions such as drilling by humans who do not know the existence of disposal facilities after the institutional control period (ICP). An inadvertent human intrusion would be the most critical factor to limit the radioactivity concentration of the repository because it is difficult to predict the future human behavior patterns and uncertainties. Therefore, the inadvertent intrusion should be considered when establishing decontamination factor (DF) targets for spent nuclear fuel reprocessing. In this paper, DFs of actinides and Sr/Cs in the spent nuclear fuels of the marine reactor, designed by the Ulsan National Institute of Science and Technology, are derived so that the radiological consequences of the inadvertent human intrusion into the waste repository are below the domestic and international criteria. Then, spent nuclear fuel (SNF) reprocessing processes, which have been operated or are under development, are investigated to confirm the feasibility of achieving the derived DFs for the marine reactor.

2. Methods and Results

In this section, the method used to calculate the dose rate of the SNF of the marine nuclear reactor, and the results are described.

2.1 Dose Calculation Method

The radiation doses of the inadvertent human intrusion for the final waste repository are calculated based on the report of Posiva [1]. The total dose rate received by the drilling worker or geologist is evaluated by considering external irradiation from the contaminated excavated sample, inadvertent ingestion, and dust inhalation. The shielding effect is not considered. The equations used for the external, ingestion and inhalation dose are as follows:

$$D_{ext} = 1.4 \cdot 10^{-13} \cdot f_1 \cdot f_2 \cdot \frac{1}{x^2} \cdot \rho \cdot V \cdot t_{exp} \cdot \sum_i (E_i \cdot S_i) \quad (1)$$

$$D_{ing} = t_{exp} \cdot m \cdot \textstyle\sum_i (I_{ing,i} \cdot S_i), \text{ and} \quad (2)$$

$$D_{inh} = t_{exp} \cdot R \cdot d \cdot \textstyle\sum_i (I_{inh,i} \cdot S_i). \quad (3)$$

where D_{ext} is effective dose equivalent from external irradiation, D_{ing} is effective dose equivalent from ingestion, D_{inh} is effective dose equivalent from inhalation, f_1 is conversion factor from exposure to effective dose equivalent, f_2 is Self-shielding factor, x is distance from source, ρ is density of sample, V is volume of sample, t_{exp} is exposure time, E_i is mean gamma energy per disintegration, S_i is average activity concentration of radionuclide i in the sample, m is intake by ingestion, $I_{ing,i}$ is dose per unit intake by ingestion of each radionuclide i, $I_{inh,i}$ is dose per unit intake by inhalation of each radionuclide i, R is respiration rate, and d is air dust concentration.

The variables in equations depend on the drilling method and they were obtained from the Posiva's report with reference to the diamond core drilling. The exposure time and distance from the excavated sample are assumed to be 1 hour and 1 meter. The volume of the excavated sample is 0.02 m^3.

The SNF inventory of the long-life small modular lead cooled fast reactor (LFR) for the naval propulsion with metal fuel included 10% zirconium (U-10%Zr) and oxide fuel (UO_2), which is calculated by Monte Carlo simulation, is utilized. The waste glass form is assumed as the final waste form with the conventional acceptable waste loading of 10 - 20 wt.% [2]

2.2 Dose for the Inadvertent Human Intrusion

The IAEA's specific safety requirements of No. SSR-5 (Disposal of Radioactive Waste) states that the dose limit for the public from all planned exposure situations is an effective dose of 1 mSv in a year. If the reasonable efforts are warranted at the stage of development of the facility to reduce the probability of intrusion, it could be available to set the dose limit as annual doses in the range 1–20 mSv [3]. Therefore, the dose criteria of this paper for the inadvertent human intrusion are set to 1 and 20 mSv.

The external, ingestion and inhalation doses from the excavated sample from U-10%Zr and UO_2 fuel are represented in Fig. 1 and Fig. 2 respectively. Both of the fuels have almost the same patterns as time passes. The total dose of both fuels lies over 10^4 mSv even after 100,000 years, and the most dominant factor is the inhalation. The external dose meets the 1 mSv criterion around 400 years and ingestion meets the criteria around 30,000 years. The institutional control periods specified in the notice of NSSC of Korea is 300 years. The reprocessing is necessary to satisfy the ICP standard.

For the inhalation, TRUs such as Am-241, Pu-238, Pu-239, and Pu-240 take the largest portion for overall periods, and the doses of Sr-90 and Cs-137 are considerable for up to 500 years. For the ingestion, the doses from Sr-90, Cs-137, and Pu-239 contribute the largest dose portion up to 500 years, and after 500 years, TRUs such as Am-241, Pu-238, Pu-239, and Pu-240 take the greatest portion. For the external irradiation, the doses from Eu-152, Eu-154, and Y-90 are dominant up to 500 years, and TRUs such as Am-241, and Pu-239 are after 500 years.

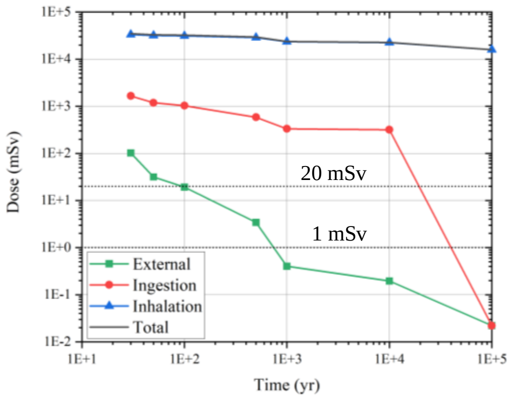

Fig. 1. Dose for the inadvertent human intrusion into the U-10%Zr SNF repository.

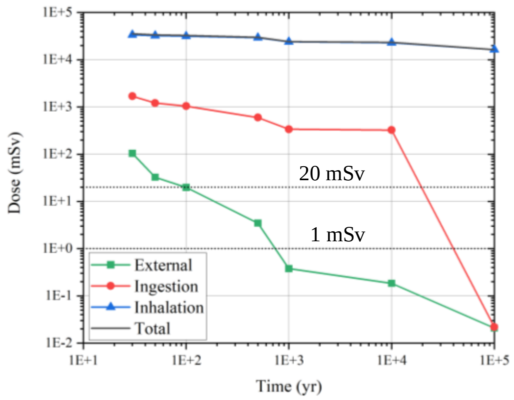

Fig. 2. Dose for the inadvertent human intrusion into the UO_2 SNF repository.

For the partitioning elements, U/TRU and Sr/Cs are considered since their radiological effects are the most principal in the long-term (after 500 years) and short-term respectively. Others are assumed as remained in the final waste.

2.3 Target Decontamination Factors

The required DFs of U/TRU and Sr/Cs to meet 1 and 20 mSv for each fuel are calculated according to the ICP and the waste loading ratio of the final waste as shown in Table I and Table II. The DFs for U/TRU and Sr/Cs are set not to exceed 100,000 and 300 by considering practically achievable DFs of the conventional reprocessing processes. The Sr/Cs decontamination coefficient is set to as small as possible.

Table I: Required decontamination factor of U/TRU and Sr/Cs according to the dose criteria for the inadvertent human intrusion into the spent nuclear fuel reprocessing final waste repository for U-10%Zr fuel

Dose criteria (mSv)		1		20	
Waste loading (wt.%)		10	20	10	20
Required DF by ICP 300 years	U/TRU	50,000	Not feasible[1]	400	800
	Sr/Cs	300		5	10
Required DF by ICP 500 years	U/TRU	10,000	40,000	200	400
	Sr/Cs	50	100	5	10
Required DF by ICP 1,000 years	U/TRU	2,500	5,000	125	250
	Sr/Cs	1	1	1	1

1: Dose for the inadvertent human intrusion dose not reached in 300 years when dose criterion is 1 mSv.

Table II: Required decontamination factor of U/TRU and Sr/Cs according to the dose criteria for the inadvertent human intrusion into the spent nuclear fuel reprocessing final waste repository for UO_2 fuel

Dose criteria (mSv)		1		20	
Waste loading (wt.%)		10	20	10	20
Required DF by ICP 300 years	U/TRU	25,000	Not feasible[1]	400	800
	Sr/Cs	300		5	10
Required DF by ICP 500 years	U/TRU	8,000	30,000	200	600
	Sr/Cs	50	100	5	5
Required DF by ICP 1,000 years	U/TRU	2,500	5,000	150	250
	Sr/Cs	1	1	1	1

1: Dose for the inadvertent human intrusion dose not reached in 300 years when dose criterion is 1 mSv.

Generally, the U-10%Zr fuel needs a slightly higher DF. If actinide elements are removed from the SNF with DF of greater than 500, the dose of the ingestion becomes more significant than that of the inhalation for the first 500 years, and the dose targets cannot be satisfied without separation of Sr/Cs. Even though all the actinides and Sr/Cs nuclides are eliminated, the final waste loaded at 20 wt.% cannot satisfy the 1 mSv and 300 years ICP due to Sm-151. In the case of 10 wt.% loaded waste, it can satisfy both dose limit and ICP standard.

2.4 Decontamination Factors of U/TRU and Sr/Cs of the Reprocessing Processes

DFs of the conventional SNF reprocessing processes and the required ICP for each process are summarized in Table III and Table IV respectively [4, 5]. Reprocessing of SNF is categorized into aqueous process and pyroprocessing.

Aqueous process is a method of dissolving spent nuclear fuel into a nitric acid solution and separating the desired nuclides using organic solvents or sedimentation agents. Four aqueous processes of PUREX, UREX+3, EURO-GANEX and 4-Group (Japan) are investigated in this study. PUREX, which is the most typical process, aims to recover only Pu and U from the spent nuclear fuel. In comparison to PUREX, UREX+3 enhances the off-gas treatment to recover I-129, H-3, and Kr-85. Also, it modifies and extends the chemical separations processes. The EURO-GANEX process was developed as part of the European Union Actinide Recycling by Separation and Transmutation and Safety of Actinide Separation processed programs. It aims to co-separate all transuranic elements from SNF without the extraction of pure Pu. The 4-Group process was developed in the Japan Atomic Energy Research Institute and for concentrated high-level liquid waste (HLLW) to separate the elements into four groups: TRU, Tc - platinum group metals (PGM), Sr/Cs and the other elements. From various experiments, the decontamination effect of 4-Group process is demonstrated. The wastes from the Rokkasho reprocessing plant can be disposed of at the low-level waste site based on radioactivity.

The pyroprocessing uses molten salt or liquid metal, not water or organic solvents and consists of electric refining, distillation, and solvent-solvent extraction. The pyroprocessing developed by Korea Atomic Energy Research Institute (KAERI) is composed of the seven processes; fuel-element chopping and decladding, high-temperature voloxidation, electrolytic reduction, electrorefining, electrowinning, salt purification and fuel fabrication. PyroGreen has been designed to convert all SNFs into low and intermediate level waste so it adopts additional processes based on the KAERI's pyroprocessing to reduce TRUs in the final wastes: a hull electrorefining, an additional actinide recovery process and Tc/I target fabrication. Through the additional processes, the DFs of the PyroGreen process is enhanced.

By adopting an additional process of removing Sr/Cs slightly, PUREX can satisfy the 20 mSv criterion within ICP of 300 years. EURO-GANEX is equivalent to PUREX but its ICP for 1 mSv criterion is 500 years unlike PUREX which is impossible within 1,000 years. Using UREX 3+ or 4-Group it is possible to meet 300 years at 20 mSv. UREX+3 needs 500 years ICP and 4-Group needs 300 years with waste loading of 10 wt.%. If the Pyroprocess of KAERI is applied, more than 10,000 years is necessary to satisfy the 1 mSv but the ICP is 300 years when the criterion is 20 mSv. PyroGreen can fulfill both dose criteria depending on the waste loading ratio.

Table III: Decontamination factors of U/TRU and Sr/Cs of reprocessing processes

Type		DF		
		U/TRU	Cs	Sr
Aqueous process	PUREX (1995+)	>1,000 Pu	N/A	N/A
	UREX+3	30,000	100	100
	EURO-GANEX	10,000	N/A	N/A
	4-Group (Japan)	50,000	10,000	10,000
Pyro-process	Pyroprocessing (KAERI)	1,000	50	100
	PyroGreen	50,000	20,000	200

Table IV: Required institutional control periods to satisfy 1 mSv and 20 mSv for the inadvertent human intrusion scenario

Type	Required ICP of U-10%Zr (y)		Required ICP of UO_2 (y)	
	1 mSv	20 mSv	1 mSv	20 mSv
PUREX (1995+)	Not feasible[1]	Within 300	Not feasible[1]	Within 300
UREX+3	500 ~ 1,000	Within 300	500 ~ 1,000	Within 300
EURO-GANEX	500 ~ 1,000	Within 300	500 ~ 1,000	Within 300
4-Group (Japan)	300 ~ 500	Within 300	300 ~ 500	Within 300
Pyro-processing (KAERI)	Not feasible[1]	Within 300	Not feasible[1]	Within 300
PyroGreen	300 ~ 500	Within 300	300 ~ 500	Within 300

1: Dose for the inadvertent human intrusion dose not reached in 1,000 years.

3. Conclusions

Decontamination factors of U/TRU and Sr/Cs for U-10%Zr and UO_2 SNF are derived from the inadvertent human intrusion analysis according to the institutional control periods by comparing the IAEA's dose criteria of 1 and 20 mSv. The dose of 1 mSv would be achievable within 300 years only in the case of 10 wt.% loading by the processes of PyroGreen and 4-GROUP. For the dose limit of 20 mSv, DFs for U/TRU of the orders of 100 and DF for Sr/Cs of about 10 are required to satisfy the ICP of 300 years. The DFs would be achievable by all the surveyed aqueous processes and pyroprocessing. Therefore, the SNFs of both U-10%Zr and UO_2 can be disposed of safely by applying the conventional or under development reprocessing processes in the aspect of the inadvertent human intrusion into the final repository.

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