CROSS-SECTORAL STUDY ON NUCLEAR ENERGY SYSTEM FOR LESS-IMPACTED RADIOACTIVE WASTE MANAGEMENT
Effect of various spent nuclear fuel properties and reprocessing conditions on geological disposal

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Abstract

In the geological disposal of high-level radioactive waste, the amount and properties of waste buried deep underground depends on various process conditions, starting with nuclear fuel. To reduce the environmental load of geological disposal, we evaluated the effects on vitrified waste heat generation of fuel burn-up, spent fuel cooling period, separation of heat-generating nuclides, and waste loading of vitrified waste, which are closely related to the design of the repository. The timing of the appearance of the maximum temperature of the buffer material in the repository depends on the heat-generating nuclides, including short-lived nuclides Cs and Sr and their daughter nuclides and long-lived minor actinides contained in vitrified waste. The generation and accumulation of these nuclides is related to the fuel burn-up and the cooling period of the spent fuel. We examined the effect of the minor actinide separation ratio, which is related to the spent fuel cooling period, on the waste-occupied area in repository to reduce the amount of vitrified waste with high loading, assuming that molybdenum and platinum group metals were separated. Based on these results, the idea of cross-sectoral and integrated research on spent fuel, nuclide separation, vitrified waste, and geological disposal was examined to present technical options that contribute to load reduction in geological disposal.

1. INTRODUCTION

Japan's strategy for nuclear energy use relies on reprocessing spent fuel, vitrifying high-level radioactive waste, and geological disposal of vitrified waste. To reduce waste volume and harmfulness, nuclide separation and transmutation technologies are being developed simultaneously. Disposal of radioactive waste is essential in using nuclear power, and if we are to continue using nuclear power, reducing the disposal load requires various issues to be considered from perspectives including technology, safety, cost, and policy.

We began a research program in FY 2014 aimed at presenting technical options that are effective countermeasures by reviewing the entire fuel cycle [1,2], in order to reduce the environmental impact of geological disposal of high-level radioactive waste and promote the use of plutonium. At present, the environmental impact of geological disposal is determined based on the amount of waste and radiation safety. We have focused on the effect of separating of minor actinides (MAs) and Cs and Sr to reduce the heat generation by vitrified waste. To reduce the disposal area required, we have also examined separating Mo and platinum group metals (PGM) to improve the vitrified waste loading, which is directly linked to the amount of glass produced. Because of the diversity of the properties of spent fuels, such as the fuel burn-up and cooling period, we compared combinations of fuel cycle conditions to reduce the area occupied by waste in the repository based on calculations.

In this paper, based on a current geological disposal system, we focus on the diversity of UO_2 spent fuel, nuclide separation, and vitrified waste with high loading. We examine the status of integrated research across the fuel cycle, which allows us to present technical options for reducing environmental load in geological disposal.

2. ENVIRONMENTAL IMPACT AND ITS REDUCTION IN GEOLOGICAL DISPOSAL

Vitrified waste is a high-level radioactive waste and is buried in repositories consisting of excavated galleries in stable deep geological formations. In Japan, it is stipulated that the waste must be buried deeper than 300 m below the surface [3]. The vitrified waste is enclosed in a metal container, overpack, and then surrounded with a buffer material mainly composed of clay and buried in the rock. There are two emplacement methods depending on whether the orientation of the waste is vertical or horizontal. The waste package pitch and the tunnel-to-tunnel distance are determined by the maximum temperature of the buffer material, which depends on the decay heat of the radionuclides contained in the glass and the mechanical stability of the tunnel excavated into the rock. These factors govern the area of the repository. Here, the maximum temperature of the buffer material was set not to exceed 100 °C to prevent alteration of the clay minerals. The product of the waste package pitch and the tunnel-to-tunnel distance is called the waste-occupied area per waste package. For a repository at a depth of 1000 m in granite with vertical emplacement, a tunnel-to-tunnel distance of 10 m, and a pitch of 4.44 m, the occupied area per waste package is 44.4 m^2 [4]. Table 1 shows the fuel cycle conditions of the vitrified waste in this reference case together with the heat generation value of the glass. The size of the repository (waste-occupied area) of the reference case is set considering the heat generation of the vitrified waste, which is determined by the reactor, fuel, and reprocessing and vitrification conditions shown in Table 1.

TABLE 1. FUEL CYCLE CONDITIONS FOR THE VITRIFIED WASTE FOR THE REFERENCE CASE

Reactor	Fuel	Burn-up (GWd/t HM)	Spent fuel cooling period (year)	Reprocessing/ Purex process/ nuclide separation (%)		Vitrification/ waste loading (wt%)	Vitrified waste			Geological disposal/waste occupied area (m^2)
							*	**	***	
LWR	UO_2	45	4	U/Pu	99.6/99.5	Approx. 20	2.3	50	0.35	44.4

* Heat generation rate, after vitrification (kW/glass unit)

** Storage period (years)

*** Heat generation rate, after storage (kW/glass unit)

The maximum estimated radiation dose that people living on the surface receive from the nuclide migration from 40 000 vitrified wastes buried in the reference case repository is 5×10^{-3} μSv/year after approximately 8×10^5 years of disposal [5]. This value is several orders of magnitude lower than the value of the dose constraint for geological disposal prescribed by safety regulations in other countries, and the main contributing nuclide is ^{135}Cs. The estimation of nuclides released from the vitrified waste into groundwater 10^7 years after disposal indicates that approximately 99% of the initial inventory of ^{237}Np remains in the engineering barrier and approximately 95% of the initial inventory of ^{135}Cs remains in the natural barriers [5]. Isolation and confinement are the fundamental concepts for securing long-term radiation safety in geological disposal.

To reduce waste volume and harmfulness, research and development of separation and transmutation technologies for long-lived and heat-generating nuclides are under way. For example, under the same fuel conditions as described above, the radiotoxicity per 1 tHM of spent fuel was compared for spent fuel, high-level waste (vitrified waste), and MA separation/transmutation waste. The time required for the radiotoxicity/ingestion in these cases to reach the same value as natural uranium was estimated at approximately 10^5, 10^4, and a few hundred years, respectively. This shows the effects of the reduction of radiotoxicity by separating 99.5% of MA from high-level waste [6].

Mitigating the heat generation of vitrified waste and reducing the amount of waste generated reduces the waste-occupied area and the entire repository area. Therefore, the amount of waste is a measure for evaluating environmental load in geological disposal. The environmental load of radiation effects can also be reduced by separating and transmuting highly radiotoxic nuclides in waste.

In this study, we calculated the effect of various fuel cycle conditions on the amount of waste, to reduce the environmental load without changing the method of geological disposal.

3. EFFECT OF FUEL CYCLE CONDITIONS ON THE REPOSITORY

In previous studies, we evaluated the reduction of the load of geological disposal by introducing the Comprehensive Analysis of Effects on Reduction of disposal Area (CAERA) evaluation index [1,7], which compares the reduction effect of the parameters of each process in the fuel cycle on the waste-occupied area per waste package in the repository. This index shows the oxide equivalent waste weight per waste-occupied area in the repository and the unit is kilograms per meters squared. The larger this value, the greater the area reduction effect on the repository. Using this evaluation index, we obtained the following effects of nuclide separation on the geological disposal of vitrified waste generated from UO_2 fuel [1].

(a) Waste-occupied area can be reduced by about 40% by producing vitrified waste with a high loading through separating the Cs and Sr heat-generating nuclides, and Mo and PGM, which affect the glass properties.
(b) Waste-occupied area can be reduced by about 40% by MA separation, owing to the relationship between increasing the spent fuel cooling period and increasing the waste loading of the glass.
(c) Nuclide separation technology can be incorporated into the fuel cycle process because the nuclide separation ratio expected to reduce the waste-occupied area is 70 to 90%.

In this paper, we examine the relationship between fuel cycle conditions and geological disposal by focusing on the burn-up of UO_2 fuel and the cooling period of the spent fuel and we discuss the temperature change of buffer materials at the repository.

3.1. Burn-up of UO_2 fuel and cooling period of spent fuel

3.1.1. Calculation conditions

ORIGEN 2.2 UPJ was used for calculating nuclide composition, decay, and reprocessing of spent fuel. ORLIBJ 40 was used as the effective cross section library, and ENSDF was used as the decay library for a 17 × 17 pressurized water reactor fuel assembly. The calculation conditions are shown in Table 2.

TABLE 2. CALCULATION CONDITIONS

Burn-up (GWd/tHM)	U-235 enrichment (wt%)	Specific power (MW/MTU)	Operation days
28	3.4		736.84
45	4.1	38	1184.21
70	4.7		1842.11

The weight of the vitrified waste was 400 kg for the reference case. The heat generation rate during production was ≦2.3 kW/glass unit. A MoO_3 content of ≦1.5 wt %, a PGM content of ≦1.25 wt %, and a Na_2O content of 10 wt % were used for the vitrified waste properties [2,8]. We assumed that the vitrified waste was stored in a surface facility for cooling for 50 years, and then buried in the repository. When the waste was placed in the repository, the heat generation rate was ≦0.35 kw/glass unit [4,5]. Based on these conditions, the waste loading of vitrified waste was adjusted according to the fuel burn-up and the cooling period of the spent fuel.

3.1.2. Vitrified waste properties

The properties of vitrified wastes corresponding to the fuels for each burn-up and cooling period are shown in Table 3. The amount of vitrified waste generated with burn-ups of 28 and 45 GWd/tHM was 1.25 glass unit/tHM, which was the same as the reference case. In contrast, 1.91 glass unit/tHM was generated for a burn-up of 70 GWd/tHM, because it exceeded the 2.3 kW/glass unit limit at the vitrified waste generation rate of 1.25 glass unit/tHM.

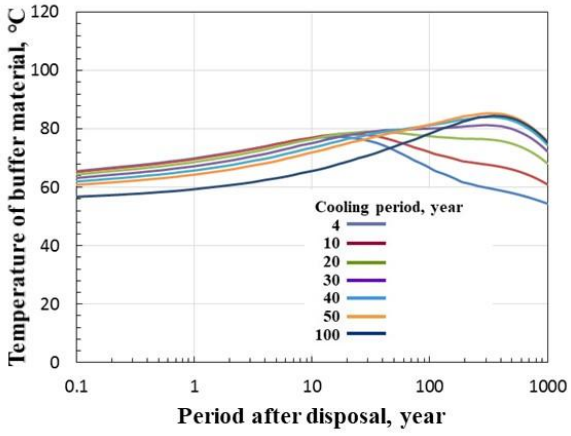
TABLE 3. PROPERTIES OF VITRIFIED WASTE

Burn-up (GWd/tHM)	Spent fuel cooling period(year)		4	10	20	30	50	100
28	Waste loading (wt%/glass unit)		17.18	17.24	17.31	17.36	17.41	17.44
	Heat generation rate (kW/glass unit)	Vitrification	1.55	0.68	0.53	0.44	0.32	0.17
		Disposal	0.216	0.213	0.204	0.191	0.166	0.122
45	Waste loading (wt%/glass unit)		20.83	20.92	21.01	21.07	21.13	21.17
	Heat generation rate (kW/glass unit)	Vitrification	2.30	1.10	0.83	0.68	0.48	0.24
		Disposal	0.343	0.328	0.301	0.273	0.225	0.155
70	Waste loading (wt%/glass unit)		20.69	20.75	20.82	20.86	20.91	20.94
	Heat generation rate (kW/glass unit)	Vitrification	2.30	1.15	0.85	0.68	0.46	0.21
		Disposal	0.348	0.325	0.288	0.254	0.201	0.128

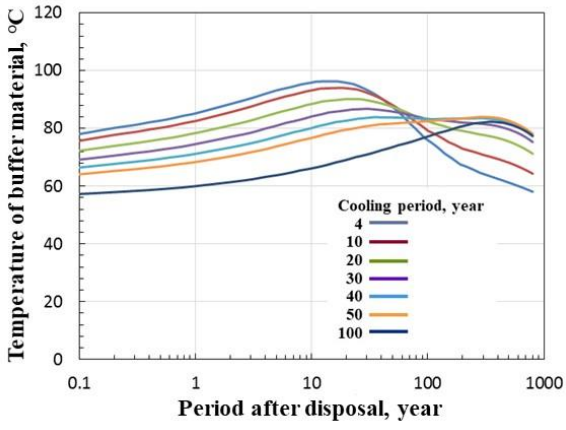
3.1.3. Geological disposal of vitrified waste

Fig. 1 (a) to (c) show the changes in the buffer material temperature when the vitrified waste for each burn-up were disposed of in the repository. Under all conditions, the maximum temperature of the buffer material was lower than 100 °C for the same waste-occupied area of 44.4 m^2, the same as the reference case. Therefore, the waste occupied area can be reduced compared with the reference case for all conditions.

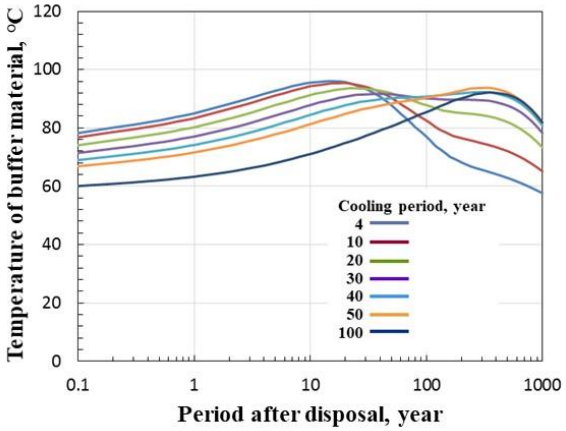
The buffer material reached the maximum temperature 20 to 30 years after disposal at any burn-up for a cooling period of 30 years or less, but when the cooling period was over 40 years, the maximum was 300 to 400 years after disposal; thus, a prolonged spent fuel cooling period delayed the time to reach at the maximum temperature. This result is explained by the decay effect of ^{90}Sr (half-life 28.8 years) and ^{137}Cs (30.2 years), and its daughter nuclides ^{90}Y (432.2 years) and ^{137m}Ba (2.55 min) in the early stage, and the production and accumulation of ^{241}Am (432.2 years) by β-decay of ^{241}Pu (14.55 days). In addition, the maximum temperature of the buffer material was higher for a lower burn-up and a prolonged spent fuel cooling period because the contribution of ^{241}Am to heat generation was larger than that of fission products (FPs) owing to the uranium enrichment of the fuel. The contributions of the main nuclides in spent fuel to heat generation are shown in Table 4.



(a) 28 GWd/tHM



(b) 45GWd/tHM



(c) 70GWd/tHM

FIG. 1. Changes in temperature of buffer materials due to heat generation of vitrified waste for different burn-ups.

TABLE 4. % CONTRIBUTIONS OF MAIN NUCLIDES TO HEAT GENERATION IN SPENT FUEL

Burn-up (GWD/tHM)	Spent fuel cooling period(year)	4	10	20	30	50	100
28	^{241}Am	1.5	6.5	12.9	18.5	28.5	49.5
	^{244}Cm	3.2	5.4	4.7	3.7	2.3	0.6
	^{90}Sr	3.1	5.8	5.8	5.4	4.4	2.3
	^{90}Y	15.0	27.8	27.7	25.6	21.1	10.9
	^{137}Cs	4.5	8.4	8.5	7.9	6.7	3.6
	^{137m}Ba	13.7	25.5	25.6	23.9	20.1	11.0
	Total (six nuclides)	41.0	79.5	85.2	85.0	83.1	77.8
45	^{241}Am	1.2	4.8	9.6	13.9	22.1	41.1
	^{244}Cm	4.1	6.4	5.7	4.6	3.0	0.8
	^{90}Sr	3.5	6.0	6.0	5.7	4.8	2.7
	^{90}Y	16.7	28.4	28.8	27.1	23.0	12.7
	^{137}Cs	4.7	8.0	8.2	7.8	6.8	3.9
	^{137m} Ba	14.1	24.2	24.8	23.6	20.5	11.9
	Total (six nuclides)	44.3	77.9	83.2	82.7	80.1	73.2
70	^{241}Am	0.9	3.5	7.0	10.3	16.6	32.4
	^{244}Cm	6.7	9.9	8.8	7.3	4.8	1.4
	^{90}Sr	3.5	5.5	5.7	5.4	4.7	2.7
	^{90}Y	16.5	26.4	27.2	25.8	22.4	13.0
	^{137}Cs	4.6	7.4	7.7	7.4	6.5	4.0
	^{137m} Ba	13.8	22.3	23.2	22.3	19.7	12.1
	Total (six nuclides)	46.0	74.9	79.6	78.4	74.7	65.5

3.2. High vitrified waste loading and separation of MA

3.2.1. Calculation conditions

The calculation of the basic properties of the vitrified waste was the same as in Section 3.1. However, to reduce the amount of vitrified waste by increasing the waste loading to up to 35 wt %, the separation ratio of MoO_3 and PGM, which affect the properties of vitrified waste, from the high-level waste liquid was set to 70%. For a prolonged spent fuel cooling period, considering the effect of ^{241}Am described in Section 3.1, the separation ratio of MA (Np, Am, and Cm) was set to 0, 70, and 90%, and the effect on the heat generation was evaluated.

3.2.2. Vitrified waste properties

The vitrified waste properties for a prolonged spent fuel cooling period at a burn-up of 45 GWd/tHM are shown in Table 5. For vitrified waste with 35 wt % waste loading, at a heat generation rate of less than 2.3 kW/glass unit as in the reference case, 70 to 90% MA separation was effective when the spent fuel cooling period was 20 years or more. It is expected that separating Cs and Sr, which are short-lived heat-generating nuclides, requires vitrified waste with up to 35 wt % waste loading for a spent fuel cooling period of 4 to 10 years.

TABLE 5. PROPERTIES OF VITRIFIED WASTE FOR EACH SPENT FUEL COOLING PERIOD AND MA SEPARATION RATIO (FUEL BURN-UP, 45 GWD/THM; WASTE LOADING, 35 WT %; AND MO/PGM SEPARATION 70%)

Spent fuel cooling period (year)	MA separation (%)	Heat generation rate (kW/glass unit)	MoO_3 content (wt%)	PGM content (wt%)	Waste generation (glass unit/tHM)
4	0	5.80	1.16	0.90	0.447
	70	5.65	1.18	0.92	0.438
	90	5.60	1.19	0.93	0.435
10	0	3.03	1.15	0.90	0.451
	70	2.80	1.18	0.92	0.439
	90	2.77	1.19	0.93	0.436
20	0	2.28	1.14	0.89	0.455
	70	2.05	1.17	0.92	0.441
	90	1.99	1.18	0.92	0.437
30	0	1.86	1.13	0.88	0.458
	70	1.63	1.17	0.91	0.442
	90	1.56	1.18	0.92	0.437
50	0	1.29	1.12	0.88	0.461
	70	1.05	1.17	0.91	0.443
	90	0.98	1.18	0.92	0.438
100	0	0.64	1.12	0.87	0.463
	70	0.40	1.16	0.91	0.444
	90	0.33	1.18	0.92	0.439

3.2.3. Geological disposal of vitrified waste

Table 6 shows the relationship between the prolonged spent fuel cooling period and the MA separation ratio, vitrified waste properties, maximum temperature of the buffer material, and waste-occupied area in the repository. When the spent fuel cooling period was over 50 years and 90% MA separation was achieved, the vitrified waste could be buried in the same area as the reference case of 44.4 m^2. For a cooling period of 30 years or more, vitrified waste with 35 wt % waste loading could be buried by increasing the waste-occupied area at 90% MA separation. These results are explained by the reduction in heat generation of Cs and Sr and the accumulation/separation of Am-241 in the prolonged spent fuel cooling period.

TABLE 6. RELATIONSHIP BETWEEN THE MAXIMUM TEMPERATURE OF THE BUFFER MATERIAL AND WASTE-OCCUPIED AREA FOR EACH SPENT FUEL COOLING PERIOD AND MA SEPARATION RATIO (FUEL BURN-UP: 45 GWD/THM; WASTE LOADING: 35 WT %; AND MO/PGM SEPARATION: 70%)

Spent fuel cooling period (year)	MA separation (%)	Max. temp. of buffer material (°C) at 44.4m^2 of waste occupied d area	Required waste occupied area at less than 100 °C of buffer material(m^2)
30	70	130.0	300 (101.3 °C)
	90	118.7	93 (99.9 °C)
50	0	181.2	300 (108.4 °C)
	70	107.7	56 (99.9 °C)
	90	93.9	-
100	0	178.3	143(99.7 °C)
	70	87.1	-
	90	65.0	-

4. CROSS-SECTORAL AND INTEGRATED RESEARCH

Based on the use of nuclear energy in the middle to the second half of the 21st century, it is expected that fuel burn-up will increase and the use of MOX fuel will expand, with prolonging the spent fuel cooling period. These parameters are set at the beginning of the fuel cycle, and they affect the properties of the vitrified waste and its geological disposal. Table 7 shows the relationship between the multiple conditions in the fuel cycle and geological disposal. Considering the conditions for disposal area reduction as for environmental load reduction in geological disposal, the heat generation rate of vitrified waste and the waste loading related to the amount of vitrified waste should be paid attention simultaneously. For example, we expect that multiple technical options for reducing the load of the repository will be presented by combining the following conditions.

Nuclear fuel: Based on the relationship between fuel burn-up, fuel composition, and spent fuel cooling period, the contribution of FPs and MA (especially ^{241}Am) to the heat generation rate of the vitrified waste can be changed.

Nuclide separation: The separation ratio between FPs (short half-life) and MA (long half-life) affects the heat generation of vitrified waste. In addition, the MoO_3 and PGM separation ratio affects the vitrified waste properties and its waste loading. To reduce the waste occupied area at the repository, we estimate that separation ratios of 70 to 90% for MA, 70 to 90% for Cs and Sr, and 70% for MoO_3 and PGM are effective.

Vitrification: Increasing the waste loading of vitrified waste reduces the amount of waste generated, although it causes an increase in the waste occupied area due to the increased heat generation rate.

Geological disposal: The waste package pitch and tunnel-to-tunnel distance must be set considering the heat generation rate of vitrified waste and the mechanical stability of the excavated cavity, which determines the waste occupied area. The entire repository area and the tunnel excavation volume are also determined by the total amount of waste. The pitch and tunnel-to-tunnel distance may vary depending on factors including the emplacement method of waste packages (vertical or horizontal), geological formation (hard or soft rock), and the depth of the repository.

TABLE 7. DIVERSITY OF FUEL CYCLE CONDITIONS LEADING TO LOAD REDUCTION FOR GEOLOGICAL DISPOSAL

						Pre-conditioning of HLLW/nuclide separation				Vitrified Waste		Geological disposal	
Reactor	Fuel	Burn-up (GWd/tHM)	Spent fuel cooling period(year)	Reprocessing/ Purex process/	nuclide separation (%)	Nuclides			SR* (%)	WL**(wt%)	SP***(year)	Emplacement	Effects on repository
LWR	UO_2	45	4	U/	99.5	Heat generation	MA	Np	0	20	<50	Vertical	Waste package pitch (m)
			15	Pu				Am	70	25	50		Tunnel-to-tunnel distance (m)
			20					Cm	90	30	>50		Waste occupied area (m²)
			30				FPs	Cs	99	35			Entire repository area (m²)
	MOX	>45	50					Sr	99.5			Horizontal	Tunnel excavation volume (m³)
			70			Property of glass	Mo	Mo	99.9				
			100				PGM	Ru	100				
								Rh					
								Pd					

*SR: Separation ratio. **WL: Waste loading. ***SP: Storage period.

To select effective technical options for load reduction in waste disposal, we need a cross-sectoral examination of the fuel cycle and evaluation indices for comparing and studying the load reduction comprehensively and quantitatively for various combined fuel cycle conditions.

5. CONCLUSIONS

We focused on the amount of waste as a target for environmental load reduction of the geological disposal for high-level radioactive waste. We examined the effect of fuel burn-up, spent fuel cooling period, waste loading of vitrified waste, and separation of MA as heat-generating nuclides (especially ^{241}Am) and discussed their relationships with the cycle conditions for UO_2 fuel. To compare and present the combinations of cycle conditions that are expected to reduce the load at the repository, evaluation indices are required that represent the values cross-sectionally and quantitatively considering various cycle conditions. Multiple technical options are expected to be available depending on the combination of fuel cycle conditions and the point of interest.

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