REDUCTION OF GEOLOGICAL DISPOSAL AREA BY INTRODUCING PARTITIONING TECHNOLOGIES UNDER CONDITIONS OF HIGH BURN-UP OPERATION AND HIGH CONTENT VITRIFIED WASTE

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Abstract

The thermal properties and amount of vitrified waste are major factors that determine the eventual disposal area of high-level radioactive waste deep underground. The effect of high burn-up operation of a light-water reactor with UO_2 fuel on the amount and thermal properties of vitrified waste under various nuclear fuel cycle conditions was discussed. In addition, the effect of Cs and Sr separation and high-content vitrified waste on reducing the waste-occupied area, which may affect the geological disposal area, under high burn-up conditions was quantitatively evaluated by using the Comprehensive Analysis of Effects on Reduction of disposal Area (CAERA) index. The fuel burn-up had a limited effect on the amount of vitrified waste. Furthermore, the contribution to the heat generation rate of vitrified waste for high burn-up conditions of ^{137}Cs, ^{90}Sr, and their daughter nuclides, which have relatively short half-lives, increased and contribution of ^{241}Am, which has a longer half-life, decreased. Therefore, high burn-up conditions reduced the waste-occupied area via Cs and Sr separation, and the maximum effect was a reduction of 74% of the waste-occupied area with a fuel burn-up of 70 GWd/tHM, 4-year spent fuel (SF) cooling period, 90% Cs and Sr separation, and 30 wt % vitrified waste loading. The results suggested that fuel burn-up, SF cooling period, partitioning technology, and vitrified waste loading are important for the geological disposal area, and it is necessary to consider the combination of these conditions for reducing the geological disposal area.

1. INTRODUCTION

According to Japan's 5th Strategic Energy Plan in 2018 [1], nuclear power generation is planned to provide the base load power supply in Japan. Japan's basic policy for nuclear energy use is based on the nuclear fuel cycle; thus, it is essential to develop and sophisticate the nuclear fuel cycle. However, when constructing the nuclear fuel cycle and sustaining nuclear energy use, it is necessary to dispose of a large amount of high-level radioactive waste (HLW). Therefore, the reducing geological disposal area and amount of HLW are required. The thermal properties and amount of vitrified waste are two major factors that determine the emplacement of the waste packages and the eventual disposal area deep underground. These two factors, in turn, depend strongly on the radionuclide inventories contained in the HLW. The inventories mainly depend on the nuclear fuel cycle conditions, which are the fuel burn-up, the spent fuel (SF) cooling period, partitioning technologies used in reprocessing, and the vitrified waste loading. Therefore, it is necessary to evaluate comprehensively and quantitatively how these conditions affect the thermal characteristics and the amount of vitrified waste generated. In particular, evaluating the effect of fuel burn-up for HLW disposal is important for improving the economic efficiency and fuel utilization rate.

In this study, we quantitatively investigated the impact on the thermal properties and amount of vitrified waste of high burn-up operation of a light-water reactor with UO2 fuel under various fuel cycle conditions. In addition, we assumed that Cs and Sr separation processes and high-content vitrified waste were used for the current nuclear fuel cycle to reduce the amount of HLW and geological disposal area. The effect of Cs and Sr separation and high-content vitrified waste under high burn-up conditions on geological disposal area was also discussed by using Comprehensive Analysis of Effects on Reduction of disposal Area (CAERA) index.

2. CALCULATION METHOD AND CONDITIONS

2.1. Fuel burn-up conditions

The reactor operating conditions are shown in Table 1. The fuel burn-up, nuclide generation in SF, decay heat generation rate, reprocessing, and partitioning were calculated with Origen 2.2-upj [2], and the cross section libraries were based on JENDL-4.0 [3]. The fuel was UO_2 and its enrichments were assumed to be 4.5 and 6.5 wt % for burn-up rates of 45 and 70 GWd/tHM, respectively. The fuel was burned assuming a burn-up rate of 38 MW/tHM for 1184 and 1842 days in a 17 × 17 pressurized water reactor assembly. Fuel shuffling in the reactor operation was not considered.

TABLE 1. CALCUATION CONDITIONS FOR REACTOR OPERATION

Reactor operation	Fuel burn-up, GWd/THM	45	70
	Specific power, MW/THM	38	
	Operation period, days	1184	1842
	Enrichment, wt%	4.5	6.5

2.2. Reprocessing and partitioning conditions

The reprocessing conditions were assumed to be those for a typical PUREX process after several years of SF cooling. During reprocessing, 99.6% of U, 99.5% of Pu, and 100% of volatile elements, such as H, C, I, Cl, and noble gases (He, Ne, Ar, Kr, Xe, and Rn) were removed (Table 2) [4]. The high-level liquid waste (HLLW) was regarded as the residue after reprocessing. The reference case and present [5] case were studied as follows and these conditions are compared in Table 3.

(1) Reference case: the SF after discharge was reprocessed after a 4-year cooling period and no partitioning.

(2) Present study: the SF was reprocessed after a 4- to 50-year SF cooling period. It was assumed that 70% of Mo and PGM (Ru, Rh, and Pd) were separated from HLLW to satisfy the upper limit of MoO_3 and PGM loading in HLW to maintain the quality of vitrified waste and stable operation of the glass melter. In addition, the Cs and Sr separation from HLLW was assumed to be 90%. The separated elements were assumed to be immobilized and disposed of deep underground. However, the disposal area was not considered.

TABLE 2. CALCULATION CONDITIONS FOR REPROCESSING AND VITRIFICATION

Reprocessing	U, %	99.6
	Pu, %	99.5
	H, C, I, Cl, Noble gas, %	100
Vitrification	Glass weight, kg/unit	400
	Na_2O content, kg/unit	10
	Heat generation rate, kW/unit	≦ 2.3
	MoO_3 content, wt%	≦ 1.50
	PGM content, wt%	≦ 1.25

TABLE 3. COMPARISON OF SETTINGS FOR REFERENCE CASE AND PRESENT CASE

Event	Factor	Reference case	Present work
Reactor operation	Fuel burn-up, GWd/THM	45	45 and 70
Reprocessing	Cooling period of SF, years	4	4 and 50
	Mo separation ratio, %	0	70
	PGM separation ratio, %	0	70
	Cs・Sr separation ratio, %	0	0 to 90
Vitrification	Waste loading, wt%	20.8	15 to 35
Geological Disposal	Waste occupied area, m^2/vitrified waste unit	41.7	13.9 to 300

2.3. Vitrification conditions

The vitrification conditions and requirements were as follows (Table 3).

(a) The weight of vitrified waste was assumed to be 400 kg.
(b) The loading of sodium oxide (Na_2O) was 10 wt % (corresponding to 40 kg) in the vitrified waste to maintain the appropriate viscosity of the melted glass in vitrification.
(c) The upper limit of the heat generation rate of the vitrified waste was assumed to be less than 2.3 kW per vitrified waste unit, consistent with the current requirement of Japan's interim storage facility [6].
(d) The upper limits of the MoO_3 and PGM loading in vitrified waste were assumed to be less than 1.5 and 1.25 wt %, respectively, to prevent yellow phase formation in the vitrified waste and deposition of PGM in the bottom nozzle of the glass melter [7-9].
(e) The vitrified waste was assumed to be stored for 50 years after vitrification to reduce the heat generation rate of vitrified waste before disposal.

2.4. Geological disposal conditions

The geological disposal site and the thermal analysis were modeled with COMSOL Multiphysics code [10]. The analytical model used horizontal emplacement of the vitrified waste in crystalline rock (hard rock), as proposed elsewhere [11]. The initial temperature, geothermal temperature gradient and thermal conductivity of the materials in the analytical model have been used elsewhere [12].

The area required for disposing of one unit of vitrified waste is called the waste-occupied area, expressed by the product of the disposal tunnel spacing (xD) and waste package pitch (y). In addition, the waste-occupied area should be set to satisfy the upper temperature limit of the bentonite buffer material. In this study, the upper temperature limit was 100 °C, as used in Japan's geological disposal programs [5], and the disposal tunnel spacing, waste package pitch, and waste-occupied area were calculated at a buffer temperature of 100 °C. The upper temperature limit of buffer, 100℃ was conservatively assumed as temperature of illitization of bentonite which is the main component of buffer. The reason is that the performances such as water sealing property and adsorption property of nuclides required for buffer are decreased by illitization. In the reference case, the waste-occupied area was 41.7 m^2/glass unit (xD of 13.3 m × y of 3.13 m) for the horizontal emplacement configuration.

2.5. CAERA index

The CAERA index was introduced to evaluate the effect of waste-occupied area reduction under various nuclear fuel cycle conditions [13, 14]. The CAERA index was defined as:

$$\mathrm{kg/m^2} = \frac{\text{Waste loading [wt \%/glass unit]} - \mathrm{Na_2O}\text{ [wt \%/glass unit]}}{\text{Waste occupied area [m}^2\text{/glass unit]}} \times \text{Weight of vitrified waste [kg]} \times \frac{1}{100} \quad (1)$$

This index has been used to evaluate the relationship between the effect of waste-occupied area reduction and partitioning technology in the nuclear fuel cycle quantitatively by comparison with a reference case [15, 16]. In this study, the CAERA index of the reference case was calculated as 1.04 kg/m^2 for a waste-occupied area of 41.7 m^2 and a vitrified waste loading of 20.8 wt %.

3. RESULTS AND DISCUSSION

3.1. Effect of high burn-up operation on the amount of vitrified waste

Fig. 1 shows the relationship between fuel burn-up and the amount of vitrified waste for 4 and 50-year SF cooling and a vitrified waste loading of 20.8 wt %. The amount of vitrified waste per ton of SF (glass unit/tHM) and fuel burn-up (glass unit/GWd) are shown in Figs. 1 (a) and (b), respectively. The amount of vitrified waste per metric ton of heavy metal increased with fuel burn-up, whereas the amount of vitrified waste per gigawatt day did not change with fuel burn-up. The same trend was reported by Inagaki et al. in a similar study [17].

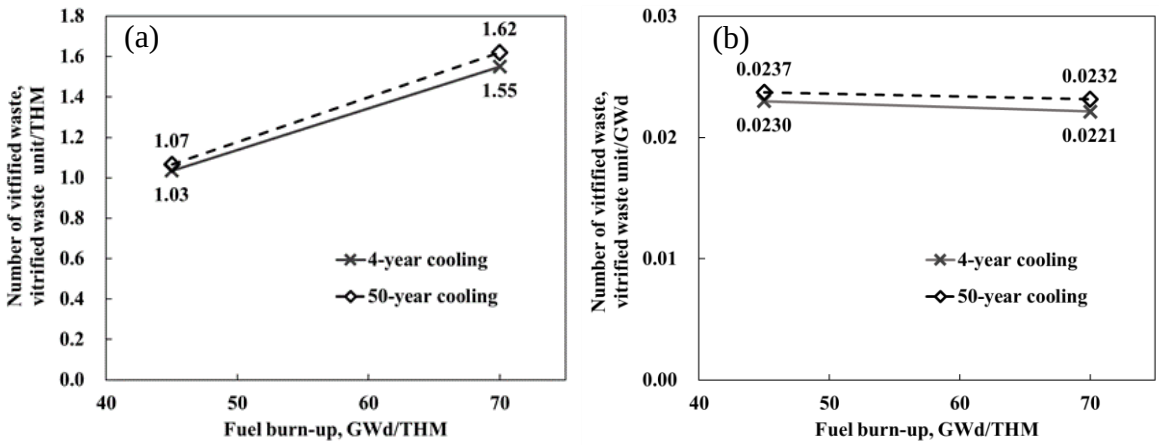


FIG. 1. Amount of vitrified waste units per (a) metric ton of heavy metal and (b) gigawatt day (SF cooling period: 4 and 50 years; waste loading: 20.8 wt %).

3.2. Effect of high burn-up operation on thermal properties of vitrified waste

Fig. 2 (a) and (b) show the heat generation of vitrified waste as a function of time from just after vitrification to disposal and after disposal, respectively, for 4- and 50-year SF cooling and 20.8 wt % waste loading. The heat generation rates of vitrified waste from just after vitrification to disposal (Fig. 2 (a)) and just after disposal (Fig. 2 (b)) were reduced by extending the SF cooling period. In contrast, more than 10 years after disposal, the heat generation rate of vitrified waste was higher for the 50-year SF cooling period than for the 4-year SF cooling period. According to Okamura et al. [15], this is because although the contribution of ^{137}Cs ($t_{1/2}$ = 30.1 years), ^{90}Sr ($t_{1/2}$ = 29.8 years), and their daughter nuclides to the heat generation rate of vitrified waste was decreased by the prolonged SF cooling period, the contribution of ^{241}Am ($t_{1/2}$ = 432 years), which has a longer half-life than ^{137}Cs and ^{90}Sr, was increased.

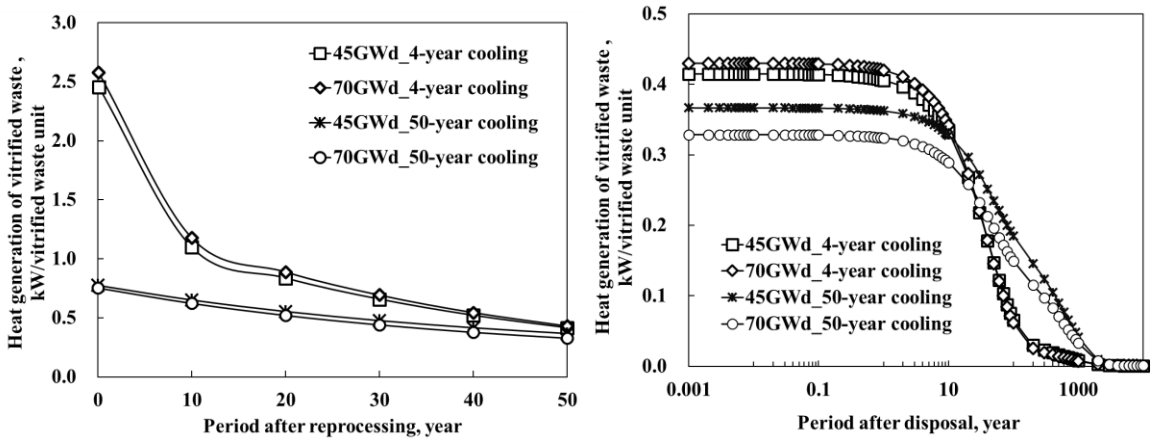
The heat generation rate was not changed by high fuel burn-up for 4-year SF cooling (Fig. 2 (b)). However, for 50-year SF cooling, the heat generation rate for a fuel burn-up of 70 GWd/tHM was lower than that for 45 GWd/tHM. Fig. 3 shows the contributions of ^{137}Cs, ^{90}Sr, ^{137m}Ba, ^{90}Y, ^{241}Am, and other nuclides to heat generation immediately after disposal (50 years after vitrification) for 4-year SF cooling. The contribution of each nuclide to heat generation was different, although the total heat generation was similar. For 70 GWd/tHM, the contribution of ^{137}Cs, ^{90}Sr, and their daughter nuclides was larger and that of ^{241}Am was smaller than for 45 GWd/tHM. Fig. 4 (a) and (b) show the contribution of each nuclide to the heat generation rate as a function of the time period after disposal at fuel burn-up rates of 45 and 70 GWd/tHM, respectively. The

contribution of each nuclide to the heat generation rate was different for each fuel burn-up rate. In addition, the contribution of ^{137}Cs, ^{90}Sr, and their daughter nuclides was still larger and that of ^{241}Am was smaller for the high burn-up conditions. Therefore, the contribution of ^{137}Cs and ^{90}Sr to the heat generation rate was higher under high burn-up conditions.

Fig. 5 shows the amount of ^{241}Am per gigawatt day in SF as a function of time after discharge from reactor for fuel burn-ups of 45 and 70 GWd/tHM. The formation path of ^{241}Am is

$$^{238}U(n,\gamma)^{239}U \rightarrow {}^{239}Np \rightarrow {}^{239}Pu(n,\gamma)^{240}Pu(n,\gamma)^{241}Pu \rightarrow {}^{241}Am \quad (2)$$

The amount of ^{241}Am was affected by ^{235}U enrichment, based on the formation path of ^{241}Am and the lower weight of both nuclides at 70 GWd/tHM than at 45 GWd/tHM. In this study, the enrichment was increased to model high burn-up. The ^{238}U content, which is the starting nuclide for ^{241}Am in fresh fuel, decreases with increasing fuel burn-up; thus, the amount of ^{241}Am should also decrease. Moreover, ^{137}Cs and ^{90}Sr, which are fission products, are produced more frequently under high burn-up conditions. Therefore, the ^{137}Cs and ^{90}Sr loading in vitrified waste increased and the contribution of these nuclides to heat generation of vitrified waste increased with fuel burn-up.



(a) Period after reprocessing until disposal *(b) Period after disposal*

FIG.2. Time course of heat generation of vitrified waste (4-year and 50-year cooling of SF, Waste loading; 20.8 wt%)

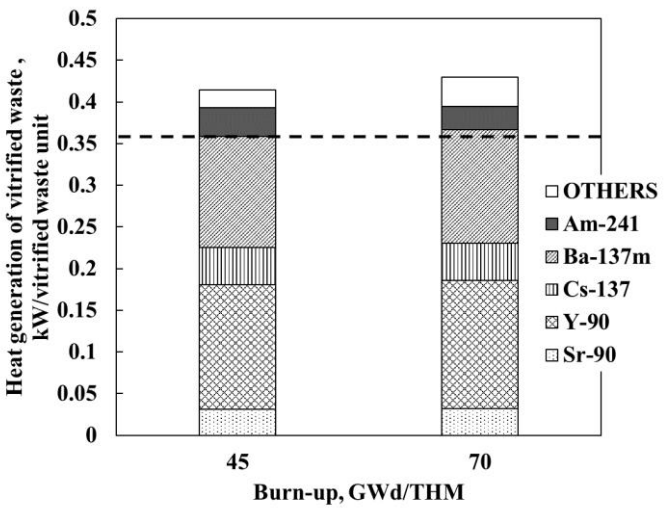


FIG. 3. Contribution of each nuclide to heat generation at disposal at fuel burn-ups of 45 and 70 GWd/tHM (SF cooling period: 4 years; waste loading: 20.8 wt %).

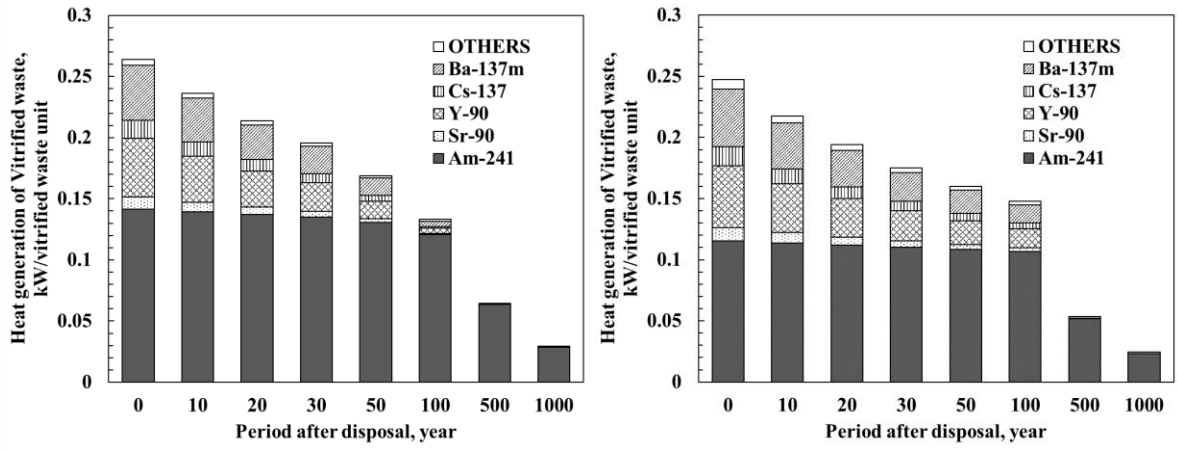


FIG. 4. Contribution of each nuclide to heat generation at fuel burn-ups of (a) 45 and (b) 70 GWd/tHM (SF cooling period: 50 years; waste loading: 20.8 wt %).

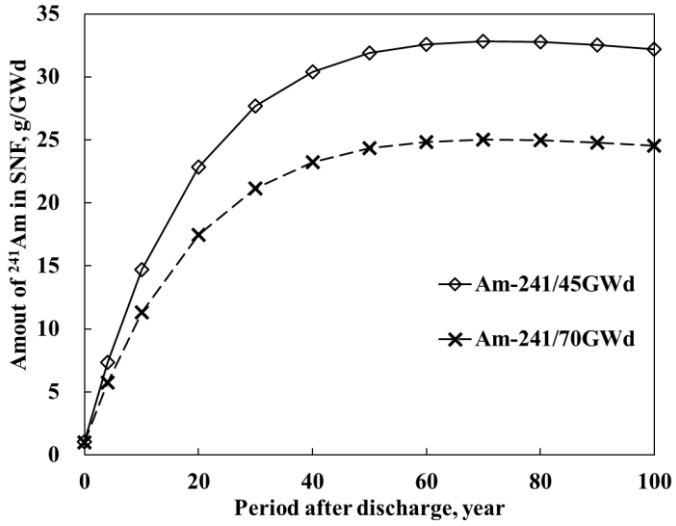


FIG. 5. Time course of amount of ^{241}Am in SF at fuel burn-ups of 45 and 70 GWd/tHM.

3.3. Effect of waste loading and Cs and Sr separation on waste-occupied area reduction

The effect of high burn-up operation on the amount of vitrified waste and thermal properties of vitrified waste is summarized as follows.

(a) The amount of vitrified waste is not changed substantially.
(b) The contribution of ^{137}Cs, ^{90}Sr, and their daughter nuclides to the heat generation rate is increased and the contribution of ^{241}Am is decreased.
(c) The contribution of ^{137}Cs, ^{90}Sr, and their daughter nuclides to the heat generation rate is reduced and the contribution of ^{241}Am is increased by extending the SF cooling period.
(d) It has been reported that the amount of vitrified waste can be reduced by increasing the vitrified waste loading [15].

Based on these results, we investigated the effect of introducing Cs and Sr separation and high-content vitrified waste on the CAERA index for high burn-up operation and shorter SF cooling period. Fig. 6 shows the relationship between vitrified waste loading and CAERA index for different fuel burn-ups for 4-year SF cooling and 90% Cs and Sr separation. The CAERA index was increased by Cs and Sr separation. For higher waste loading than the reference case, a substantially higher CAERA index was obtained. High burn-up operation at a burn-up of 70 GWd/tHM had a larger CAERA index than at 45 GWd/tHM. A maximum CAERA index of 4.00 kg/m^2 was achieved for a fuel burn-up of 70 GWd/tHM and 30 wt % vitrified waste loading. This CAERA index value indicates an approximate reduction of 74% in waste-occupied area compared with the reference case

(CAERA index = 1.04 kg/m^2). Therefore, high burn-up operation is effective for reducing the waste-occupied area by combining suitable Cs and Sr separation, SF cooling period, and vitrified waste loading conditions.

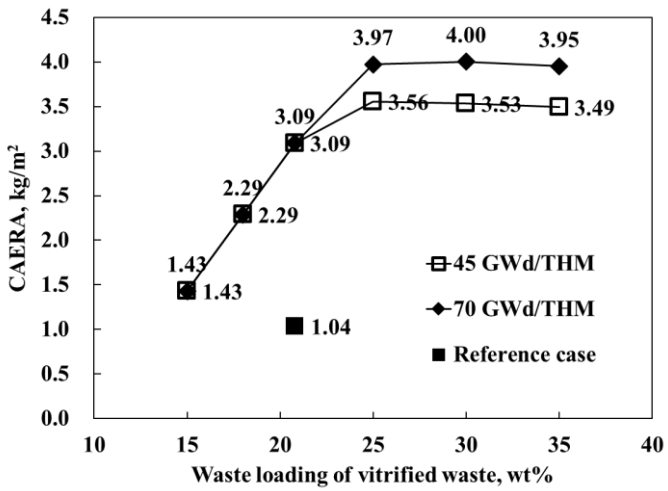


FIG. 6. Effect of Cs and Sr separation and high waste loading on CAERA index for fuel burn-ups of 45 and 70 GWd/tHM (SF cooling period; 4 years; Cs and Sr separation ratio: 90%).

4. SUMMARY

In this study, we analyzed the effect of high burn-up operation on the amount and thermal properties of vitrified waste. In addition, the effect of introducing Cs and Sr separation and high-content vitrified waste for reducing waste-occupied area was quantitatively evaluated by CAERA index. The conclusions can be summarized as follows.

(a) Increasing the fuel burn-up did not change the vitrified waste per fuel burn-up substantially, although the amount of vitrified waste per ton of SF increased.
(b) High burn-up operation increased the contribution of ^{137}Cs, ^{90}Sr, ^{137m}Ba, and ^{90}Y, and decreased the contribution of ^{241}Am to the heat generation rate of vitrified waste.
(c) Increasing the SF cooling period decreased the contribution of ^{137}Cs, ^{90}Sr, and their daughter nuclides and increased the contribution of ^{241}Am to the heat generation rate of vitrified waste.
(d) The waste-occupied area could be reduced by introducing Cs and Sr separation and high-content vitrified waste under high burn-up operation.
(e) Based on results (1)–(4), a maximum reduction in waste-occupied area of 74% could be achieved with 90% Cs and Sr separation, 30 wt % waste loading, and 4-year SF cooling.

Therefore, for high burn-up operation, it is possible to reduce the amount of vitrified waste and geological disposal area by combining suitable SF cooling period, partitioning technology, and vitrified waste loading.

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