Optimal waste loading in high-level nuclear waste glass from high-burnup spent fuel for waste volume and geological disposal footprint reduction

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Abstract

The effects of waste loading in vitrified high-level nuclear waste (HLW) from the reprocessing of high-burnup spent fuel (56 GWd/tHM) on waste volume reduction (i.e., number of HLW canisters produced) and decay heat generation were investigated to minimize the repository footprint. As the waste loading increases, the number of canisters produced decreases; however, the decay heat and subsequent footprint per canister increase. The best estimate waste loading observed was 23 wt% (including 10 wt% Na₂O), wherein the repository footprint minimizes to 60.0 m²/tHM. These results are slightly higher than those for standard HLW (55.5 m²/tHM). However, upon comparing the repository footprint with electric power generation, the benefit of optimization was that the footprint for high-burnup HLW (131 m²/TWh) was 13% smaller than that of standard HLW (151 m²/TWh).

Introduction

In view of the safe geological disposal of high-level waste (HLW) in Japan, the properties of standard vitrified HLW in canisters are evaluated assuming a spent fuel burnup of 45 GWd/tHM (Giga Watt-days per metric ton of heavy metal), a spent fuel cooling time of 4 years, and a waste loading of 20.8 wt% (including Na₂O) [1, 2]. However, considering current consumption trends, where nuclear fuel burnup continues to increase owing to the economical operation of nuclear power plants, the rise in nuclear waste inventory, decay heat, and subsequent waste volume (the number of vitrified HLW canisters produced) poses a challenge with regard to its reprocessing, interim storage, and final disposal.

By increasing the waste loading in the vitrified HLW, the waste volume can be reduced, but the decay heat generation

of the vitrified HLW canister will increase along with an increase in molybdenum and platinum group metals, which negatively affects vitrification [3]. The increase in decay heat generation affects the thermal constraint of the repository, where the temperature of the bentonite buffer surrounding the waste form is limited to below 100 °C; otherwise, the repository footprint would be greater than expected.

Accordingly, this study investigated the effects of waste loading on the waste volume and heat generation in the vitrified HLW from the reprocessing of high-burnup spent fuel and optimized the waste loading to minimize the repository footprint under the thermal constraint of the repository. We also focused on the spent fuel cooling time (interim storage period) because it is an important factor affecting the decay heat of HLW due to the build-up of ²⁴¹Am from ²⁴¹Pu decay in spent fuel.

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Methodology

The radionuclide inventory and decay heat of the vitrified HLW in the canisters were calculated using the general isotope calculation code ORIGEN2.2-UPJ [4] with the cross-section library JENDL4.0/ORLIBJ40 [5]. Based on the recent fuel developments, the spent fuel burnup is assumed to be 56 GWd/tHM. The spent fuel cooling time implies

the interim storage period before reprocessing. Recently, the shortest cooling time in the new reprocessing standard in Japan was changed from 4 to 15 years [6]. Thus, this study assumed a cooling time of 15–40 years. Detailed calculation conditions for the vitrified HLW are given in the Supplementary information (Table S1).

COMSOL Multiphysics® v. 5.6 was used for the thermal analysis of the geological repository system, to study the heat released by the vitrified HLW. The engineered barrier design, near-field system, and repository environment are based on the reference case in the Japanese safety case [1] as follows: hard rock (crystalline), 1000-m-deep underground, initial surrounding temperature of 45 °C, and vertical HLW emplacement. The boundary conditions for the heat transfer were 15 °C at the ground surface, 51 °C at the bottom (1200 m) with a temperature gradient of 3 °C/100 m, and adiabatic on all sides. Owing to the mechanical stability of the host rock, the minimum distance between the tunnels is 10 m, which is assumed to be constant in the present work. The canister pitch (distance between the vitrified HLW for vertical emplacement) is 4.44 m or higher. By increasing the pitch, the canister's footprint (area around one HLW canister) can be increased to 44.4 m²/canister or larger.

Results and discussion

Table 1 shows the effects of waste loading and spent fuel cooling time for vitrified HLW in the canisters on waste volume reduction (i.e., the number of canisters produced) and the glass components for a high-burnup HLW of 56 GWd/tHM compared with the standard HLW, which is denoted by an asterisk (45 GWd/tHM and a 4-year cooling time) [1]. As the waste loading increases, the number of canisters produced (per tons HM) can be decreased to less than 1.25 canisters/tHM for the standard HLW (20.8 wt% waste loading). The cooling time has a negligible effect on the number of canisters produced, as well as Mo and Pt group metals (PGM: Ru, Rh, and Pd). As the waste loading increases, the initial heat generation also increases but is lower than the required heat limit of 2.3 kW/canister because of the longer cooling time of 15 years or more (i.e., decay of fission products). This is also accompanied by a higher enrichment of molybdenum oxide and PGM as compared to the standard HLW. Inagaki et al. [3] suggested that the MoO₃ and PGM contents in glass are less than 1.50 wt% and 1.25 wt%, respectively. Because the

Table 1 Properties of the vitrified HLW from 56 GWd/tHM spent fuel as functions of waste loading and cooling time

Property	Cooling time (years)	Waste loading (wt%)			
		20	20.8	25	30
Number of canisters produced (canisters/tHM)	4 (45 GWd/tHM)	1.35	1.25*	0.897	0.673
	4	1.64	1.52	1.10	0.822
	15	1.68	1.55	1.12	0.838
	20	1.68	1.56	1.12	0.841
	30	1.69	1.57	1.13	0.845
	40	1.70	1.57	1.13	0.848
Initial heat generation (kW/canister)	4 (45 GWd/tHM)	2.16	2.34*	3.25	4.33
	4	2.28	2.46	3.41	4.55
	15	0.902	0.974	1.35	1.80
	20	0.796	0.860	1.19	1.59
	30	0.644	0.695	0.965	1.29
	40	0.529	0.571	0.793	1.06
MoO ₃ content (wt%)	4 (45 GWd/tHM)	1.28	1.38*	1.29	2.56
	4	1.30	1.40	1.94	2.59
	15	1.27	1.37	1.90	2.54
	20	1.26	1.37	1.90	2.53
	30	1.26	1.36	1.89	2.52
	40	1.25	1.35	1.88	2.51
PGM content (wt%)	4 (45 GWd/tHM)	1.00	1.08*	1.50	2.01
	4	1.08	1.17	1.62	2.16
	15	1.06	1.14	1.59	2.11
	20	1.05	1.14	1.58	2.11
	30	1.05	1.13	1.57	2.10
	40	1.05	1.13	1.57	2.09

*Standard HLW (45 GWd/tHM) [1]

increase in Mo and PGM is a technical challenge to be overcome in the future, this study did not consider the Mo and PGM contents in the vitrified HLW.

Figure 1a shows the post-disposal heat generation for the 56-GWd/tHM vitrified HLW and a 25 wt% waste loading (including 10 wt% sodium oxide) that had been cooled for 50 years after vitrification. The dashed line at 0.35 kW/canister is the reference level for the standard HLW at the time of disposal [1]. As mentioned previously, long-term cooling, such as 40 years, decreases the decay heat at the beginning. However, the long-term heat generation after approximately 50 years of disposal slightly increases with the 40-year cooled vitrified waste of before reprocessing as compared to the short-term cooled waste. This is because of the ^{241}Am build-up from ^{241}Pu during the interim storage cooling time.

The thermal effect of the ^{241}Am build-up in the vitrified HLW on the thermal constraint of the bentonite buffer in the repository is clearly demonstrated in Fig. 1b, where the canister's footprint is a minimum of 44.4 m²/canister. The temperature of the bentonite buffer at the hottest part of the bentonite was over the 100-°C limit in all cases. In the short-term cooling of 15 years, the temperature quickly decreased below 100 °C within 100 years. However, for the long-term cooling of 40 years, the bentonite buffer will be kept above 100 °C for nearly 900 years, leading to a large impact on the bentonite degradation to illite.

To lower the temperature of the bentonite buffer, the canister's footprint was increased beyond 44.4 m²/canister by increasing the canister pitch. According to the thermal analysis (see Supplementary information, Fig. S1), as the canister's footprint increased, the maximum temperature of the bentonite buffer in each footprint decreased. The canister's footprint, adjusted to just below 100 °C, can be obtained for the vitrified HLW with various waste loadings by tuning the pitch in detail. In the case of 25 wt% waste loading (56 GWd/tHM), the canister's footprint just below 100 °C for a cooling time of 15, 20, 30, and 40 years are

67, 63, 56, and 54 m²/canister, respectively (see Supplementary information, Table S2).

Although the canister's footprint increases for higher burnup and waste loading, the waste volume (i.e., number of canisters produced) is reduced with waste loading, as shown in Table 1. Therefore, the total repository footprint can be obtained as follows:

$$A = a \times n,$$

where A is the repository footprint (m²/tHM), a is the canister's footprint (m²/canister), and n is the number of canisters (canisters/tHM). Note that the canister's footprint is adjusted such that the minimum value is 44.4 m²/canister and the maximum temperature of the bentonite buffer will be just below 100 °C.

Figure 2a, b shows the repository footprint of the vitrified HLW for a burnup of 45 and 56 GWd/tHM with that of the standard HLW in the safety case (reference case) [1]. Regardless of the burnup, the repository footprint decreases with waste loading and then increases. The higher the waste loading, the greater the effect of cooling time. The best estimated waste loading is 23 wt%, where the repository footprint for 40-year cooled waste is a minimum of 49.1 and 60.0 m²/tHM for a burnup of 45 and 56 GWd/tHM, respectively. The canister's footprint in both cases is 46.0 m²/canister, and the number of canisters produced is 1.07 and 1.30 canisters/tHM, respectively (see Supplementary information, Tables S2, S3, S5).

Despite optimizing the waste loading, a minimum footprint of 60.0 m²/tHM for a high burnup is still larger than that for the standard HLW (55.5 m²/tHM) [1]. Additionally, the benefit of a higher burnup, that is, electricity generation, was studied. Assuming a thermal efficiency of 34% for nuclear power generation, the quantity per tHM can be converted into per electricity generation (watt-hour). The repository footprint per tera watt-hour (TWh) is shown in Fig. 2c, d, and the results are summarized in

Fig. 1 Effect of spent fuel cooling time on the time evolution of **a** heat generation rate and **b** temperature in bentonite buffer after disposal for the vitrified HLW [56 GWd/tHM and 25 wt% waste loading (including 10 wt% of Na₂O)]

Fig. 2 Repository footprint of the vitrified HLW as a function of waste loading for different cooling times. **a, b** Footprint per tons of heavy metal (tHM), **c, d** footprint per electricity generation (tera watt-hour, TWh). **a, c** 45 GWd/tHM, **b, d** 56 GWd/tHM

Table 2 Summary of the minimum footprint and optimized waste loading

	Standard HLW [1]	Present study	
Burnup (GWd/tHM)	45	45	56
Cooling time (years)	4	40	40
Waste loading (wt%)	20.8	23	23
Number of canisters produced (canisters/tHM)	1.25	1.07	1.30
(canisters/TWh)	3.40	2.91	2.85
Repository footprint (m ² /tHM)	55.5	49.1	60.0
(m ² /TWh)	151	134	131

Table 2 (all data are available in the Supplementary information, Tables S2–S6). The best estimated waste loading was 23 wt%. The number of canisters produced was reduced by 16% from 3.40 canisters/TWh for the standard HLW to 2.85 canisters/TWh. The minimum footprint for a burnup of 56 GWd/tHM was found to be 131 m²/TWh, which was 13% smaller than that for the standard HLW (151 m²/TWh) [1].

Conclusions

The waste loading for high-burnup vitrified HLW was optimized under the thermal constraint of the repository to reduce the waste volume and minimize the repository footprint. By appreciating electricity generation as a benefit of higher burnup, the best estimated waste loading was found to be 23 wt% (including 10 wt% Na₂O), where the number of canisters produced and the repository footprint can be reduced by 16% and 13%, respectively, compared to those for the standard HLW in the safety case in Japan. The increase in Mo and PGM due to the higher waste loadings poses future technical challenges for waste management strategies to reduce waste volume. There are still various conditions and uncertainties in the nuclear fuel cycle, especially for the geological environment and repository design. Further investigations are required for a better understanding of cross-cutting issues in the nuclear fuel cycle and further optimization.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1557/s43580-022-00232-3>.

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References

1. Japan Nuclear Cycle Development Institute (JNC), Second progress report on Research and Development for the geological disposal of HLW in Japan (2000)
2. The nuclear waste management organization of Japan (NUMO), NUMO safety case, NUMO-TR-20-03 (2021) (**in Japanese**)
3. Y. Inagaki, T. Iwasaki, S. Sato, T. Ohe, K. Kato, S. Torikai, Y. Niibori, S. Nagasaki, K. Kitayama, J. Nucl. Sci. Technol. **46**, 677 (2009)
4. <http://www.oecd-nea.org/tools/abstract/detail/nea-1642>
5. K. Okumura, K. Sugino, K. Kojima et al., *A Set of ORIGEN2 Cross Section Libraries Based on JENDL4.0: ORILIBJ40* (JAEA, Ibaraki, 2013)
6. Japan Nuclear Fuel Limited (JNFL), Partial amendments in the application for designation and authorization of reprocessing business (2018) (**in Japanese**)