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Evaluation of Long-Term Irradiation Field in Geological Disposal of High-Level Radioactive Wastes

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Vitrified high-level radioactive waste (HLW) is subject to alpha, beta, gamma and neutron irradiation as a result of radionuclide-decay. Radiation can cause chemical and physical effects on HLW geological disposal system, in particular, engineered barrier system (EBS) which consists of vitrified waste, overpack container and surrounded buffer material. Alpha and beta radiation can be shielded completely by the overpack as long as it retains its containment function. Gamma and neutron radiation, on the other hand, will penetrate the overpack, and then enter the buffer material and the host rock. To assess radiation effects within the EBS for long time, it is essential to evaluate the evolution of irradiation field, quantitatively. Thus, radiation transport calculations were done to obtain dose rate, irradiation dose and absorbed dose in the irradiation field. In these calculations, vitrified waste, overpack, buffer and host rock were modeled with a same concentric cylinder.

KEYWORDS: High level radioactive waste, engineered barrier system, overpack, buffer material, host rock, dose rate, irradiation dose, absorbed dose, radiation transport, Monte Carlo calculation, MCNP4A

I. Introduction

The performance of the engineered barrier system (EBS) should be evaluated comprehensively and qualitatively, taking various phenomena, which could affect the EBS, into consideration. In this evaluation, besides directly analyzing information on the irradiation field and the effects of model parameters, the features of various phenomena affecting the irradiation field and model parameters should also be trended over a long period of time. In evaluating the features of such phenomena, it is necessary to examine and pigeonhole the phenomena beforehand, clarify the interrelationship, the features and the evaluation methods of the phenomena, and extract phenomena which are considered important based on assessment of significance of effect. In the conventional evaluation of the performance of EBS, only information on the irradiation field and model parameters, which are directly used in analyzing the hydraulic features and the solubility of groundwater affected by the EBS have been taken into consideration, and discussion and analysis of various phenomena affecting the irradiation fields and model parameters has not been done in detail.

In past domestic and foreign researches on the performance of EBS, it was pointed out that the effect of radiation from vitrified HLW on the geochemistry of groundwater, which could affect the solubility and other characteristics, should be taken into consideration. Furthermore, it has been also noticed that the characteristics of the irradiation field are important factors in view of radiological protection in operation of the disposal facilities. Thus, radiation transport calculations were done to obtain dose rate, irradiation dose and absorbed dose in the irradiation field.

II. Calculation

1. Method

Neutron and gamma transport calculations were performed by using MCNP-4A of Monte Carlo code[1] and ENDF/B-5 as a nuclear data. In analysis of alpha particle, which must be taken into consideration in effect analysis of radiolysis of groundwater, it was assumed that the alpha particle from alpha emitting actinide isotope existing in the layer up to the depth of the alpha particle average range contribute to radiolysis of water. Because radiolysis by beta particle will be smaller than alpha particle on and after 500 years, analysis of beta particle was not performed.

2. Modeling

[Source Term Modeling]

Concerning about parameter to evaluate source term of vitrified HLW, 45,000 and 55,000 MWD/MTU of burn-up, 4.5 and 5.5% of U-235 enrichment and 30, 40 and 50 years as the cooling time from vitrification were used. Following four kind of source term were obtained by utilizing the output of Origen-2[2].

(1) Neutron source term by (alpha, n) Reaction
(2) A Neutron source term of Spontaneous Fission
(3) Gamma source term by Fission Products
(4) Alpha source term (not performed by Monte Carlo Calculation)

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Table 1 Calculation Conditions

Burn Up	45000, 55000 [MWD/MTU]
U-235 Enrichment	4.5, 5.5 [%]
Overpack / Thickness	0, 10, 15, 20, 25, 30 [cm]
Overpack / Material	Carbon-steel, Carbon-steel & Ti, Carbon-steel & Cu
Buffer Material / Thickness	67, 98[cm]
Buffer Material / Mixture of Quartz	0, 30 [Volume%]
Buffer Material / Water Content	0% (Operation), 100% (0,100, 1000, 10000, 100000 years after vitrified)
Cooling Time	30, 50 [years]
Elapsed Time from after Vitrification	0, 30, 50, 100, 1000, 2000, 10000, 100000 [years]
Evaluation Point	On the Vitrified HLW (side & bottom surface) On the Overpack (side & bottom surface) On the Buffer Material (side & bottom surface)

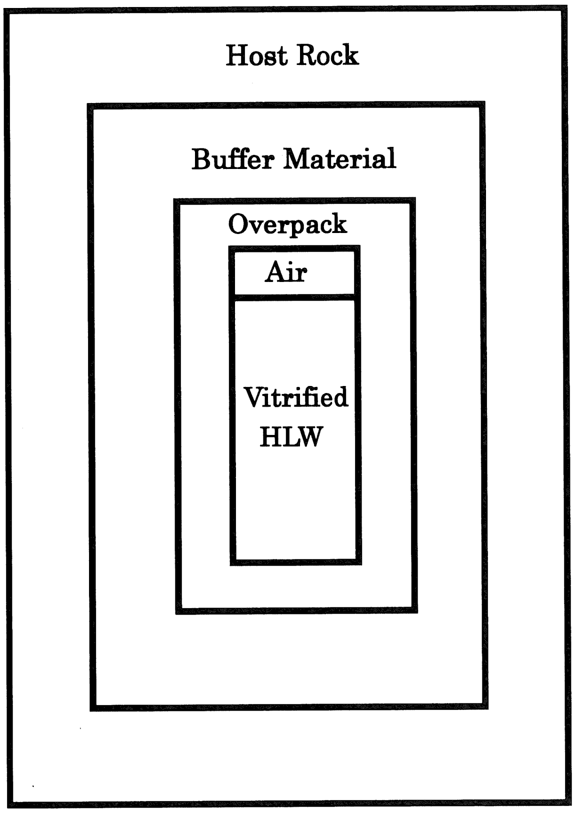

Fig. 1 Example of Calculational Geometry for Model (3)

Neutron energy spectrum and source strength by (alpha, n) reaction, and neutron strength of spontaneous fission were calculated following the procedure described on reference(3). Neutron energy spectrum of spontaneous fission were used one of Cm-244 spontaneous fission.

[Geometry]
Following three kind of models were used to radiation flux.
(1) Vitrified HLW
(2) Vitrified HLW + Overpack
(3) Vitrified HLW + Overpack + Buffered Material + Host Rock
The overpack was assumed to be made of carbon-steel, and more two kind of composite overpack coating with Ti or Cu outside of the carbon-steel, were assumed. Thickness of overpack were assumed to be ranged from 0 to 30cm. As water saturation of buffer, 0, 50 and 100 Volume% are taken into consideration. Quartz were mixtured in buffer material at the rate of 0 and 30 volume%. Calculational conditions and geometry were shown in **Table 1** and **Fig. 1**.

III. Calculation Results and Evaluation

About 25,000 cases of Monte Carlo calculations were performed. Because all cases can not be informed on this report, summary of significant results and evaluation were described in this chapter.

1. Shielding Performance of Overpack

Analysis of vitrified HLW was basis of shielding performance from the view point of radiation protect. Following calculation results, dose rate after 2000 years vitrified was about 50 mSv/h at surface of vitrified HLW, and 1m dose were about 3 mSv/h. These results introduce the necessity of overpack as shielding on transportation and operating disposal. Dose rate on surface of vitrified HLW are shown in **Table 2**, compared with the result of vitrified HLW with overpack. Table 2 shows that overpack is effective on shielding performanse, especially on gamma-ray.

2. Relation between Overpack Thickness and Dose Rate

At first, between neutron dose and gamma-ray dose are significant on shielding performance of overpack. Gamma-ray dose rate due to fission products has the highest dose rate at the overpack surface. The portion of neutron and gamma-ray dose rate contributed to total dose rate at the surface of overpack depend on thickness of ovaerpack (shown in **Table 3**). Overpack was made of carbon-steel and more effective on the attenuation of gamma-ray rather than neutron.

Next, in the case that gamma-ray dose rate was dominant, at the bottom surface was the highest compared with top and side surface. On the other hand, in the case that neutron dose rate was dominant, at the side surface was the highest compared with top and bottom surface. (following the relation between **Table 3** and **4**).

Table 2 Shielding Performance of Overpack

Elapsed time from Vitrification: 30, 100, 1000, 2000 [Years] Burn Up: 45000 MWD/MTU
U-235 Enrichment: 4.5% Overpack / Thickness: 0(Model①) or 30cm(Model②)
Buffer Material / None
Calculation Point: On the Vitrified HLW (side surface)

Dose Rate [mSv/h]	Distance from suface of Vitried HLW [cm]		Ratio
Elapsed Time from Vitrification	0cm	30cm	30cm / 0cm
30 [Years]	9.886E+05	2.157E+00	0.0002%
100 [Years]	6.864E+04	8.601E-01	0.0013%
1000 [Years]	6.053E+01	1.891E-01	0.3124%
2000 [Years]	5.139E+01	1.193E-01	0.2321%

Table 3 Relation with Overpack Thickness and Dose Rate [mSv/h]

Elapsed Time from Vitrification: 30 [Yeras] Burn Up: 45,000 [MWD/MTU] U-235 enrichment: 4.5 [%]
Calculation point: Overpack surface and 1m from overpack surface

	Overpack thickness				
Surface dose rate	10cm	15cm	17cm	20cm	25cm
TOP	5.812E+02	3.956E+01	1.467E+01	4.247E+00	1.662E+00
SIDE	6.536E+02	3.937E+01	1.554E+01	5.896E+00	2.964E+00
BOTTOM	7.947E+02	4.744E+01	1.843E+01	6.335E+00	2.593E+00
SIDE / BOTTOM	82.24%	82.99%	84.32%	93.07%	114.31%

1m dose rate	10cm	15cm	17cm	20cm	25cm
TOP	3.743E+02	2.288E+00	9.195E-01	2.761E-01	1.111E-01
SIDE	9.078E+02	5.961E+00	2.319E+00	8.214E-01	4.008E-01
BOTTOM	5.316E+02	3.707E+00	1.356E+00	4.015E-01	1.701E-01
SIDE / BOTTOM	170.77%	160.80%	171.02%	204.58%	235.63%

Table 4 Ratio of Neutron and Gamma Dose Rate [mSv/h]

Cooling Time / Vitrified: 30 [Yeras] Burn Up: 45,000 [MWD/MTU] U-235 enrichment: 4.5 [%]
Evaluation point: suface of overpack

	Overpack thickness				
Surface dose rate	10cm	15cm	17cm	20cm	25cm
Neutron Dose Rate	6.858E+00	4.947E+00	4.383E+00	3.673E+00	2.780E+00
Gamma-ray Dose Rate	6.467E+02	3.443E+01	1.116E+01	2.222E+00	1.842E-01
Neutron / Gamma	1.06%	14.37%	39.27%	165.30%	1509.23%

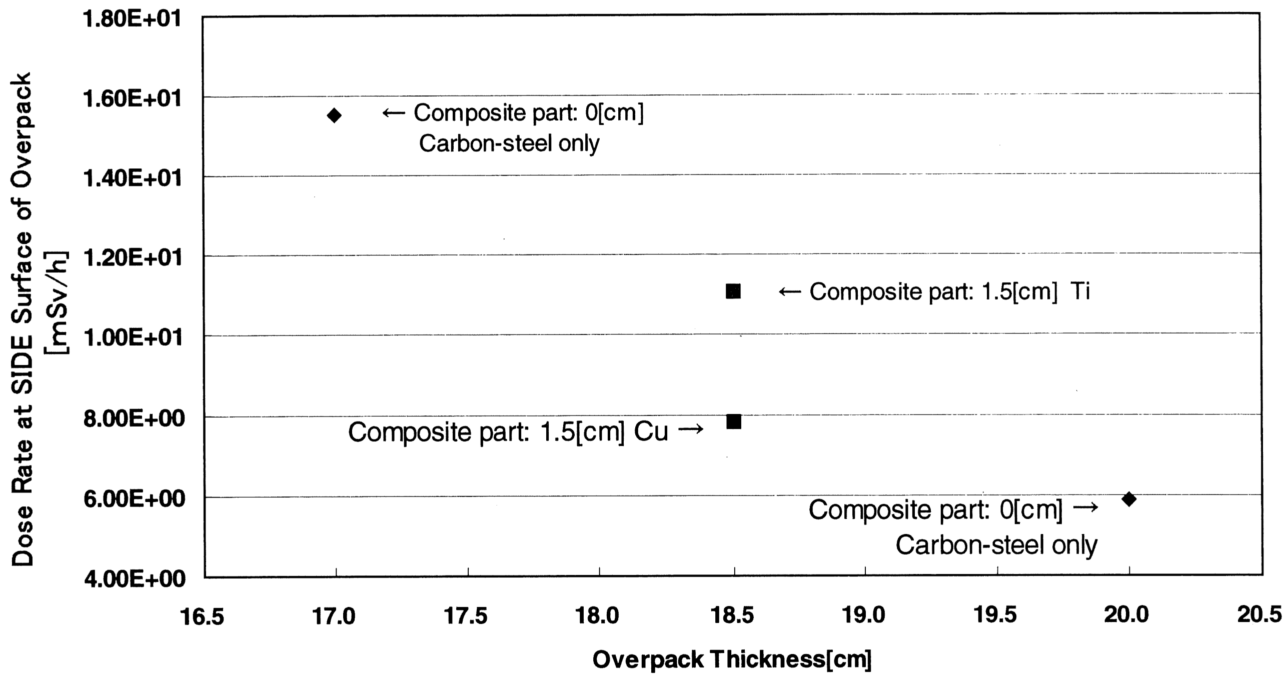

Fig. 2 Effect of Composite Overpack to Dose Rate

Table 5 Calculated Absorbed dose rate on Corrosion Products of Overpack [mGy/h]

Elapsed Time from Vitrification: 30 [Yeras] Burn Up: 45,000 [MWD/MTU] U-235 enrichment: 4.5 [%]
Evaluation point: surface of overpack overpack thickness: 17cm
Buffer Material / Mixture of Quartz: 30[Volume%] Buffer Material / Water Content: 100[Volume%]
Corrosion product of overpack was deposited ino the buffer material: 0, 30[Volume%]

Absorbed Dose Rate [mGy/h]	No corrosion product 0 [Volume%]	corrosion product 30 [Volume%]	Ratio 30 / 0
Neutron - (Alpha, n) Reaction	3.202E-02	9.712E-03	30.33%
Neutron - Spontaneous Fission	4.240E-02	1.265E-02	29.83%
Neutron - TOTAL	7.442E-02	2.236E-02	30.04%
Secondary Gamma - (Alpha, n) Reaction	6.173E-02	7.015E-02	113.63%
Secondary Gamma - Spontaneous Fission	7.655E-02	8.286E-02	108.24%
Primary Gamma - Fission Product	1.709E+01	2.955E+01	172.91%
Gamma - TOTAL	1.723E+01	2.970E+01	172.41%
Absorbed Dose Rate - TOTAL	1.730E+01	2.972E+01	171.80%

3. Effect of Composite Overpack to Dose Rate

This study was performed to understand the shielding performance of composite overpack of Ti or Cu. Thickness of carbon-steel is 17 or 20cm, and thickness of composite part is 1.5cm. Following these calculation results, in the case of Ti composite part and carbon-steel 17cm thickness, dose rate was decreased about 30~35% compared with the case of 17cm carbon-steel. In the case of Cu composite part and carbon-steel 17cm thickness, dose rate was decreased about 50~60% compared with the case of 17cm carbon-steel(shown in **Fig. 2**). Then, shielding performance is relation to order of density of each element, Cu, Fe and Ti.

4. Absorbed Dose Rate on Corrosion Products

In this study, simulation was performed to evaluate absorbed dose rate on the corrosion product of overpack deposited in the buffer material. Main material of corrosion product was made of Fe_3O_4 and diffusible range was assumed until 8.8[cm] from the surface of overpack. As shown in **Table 5**, if corrosion

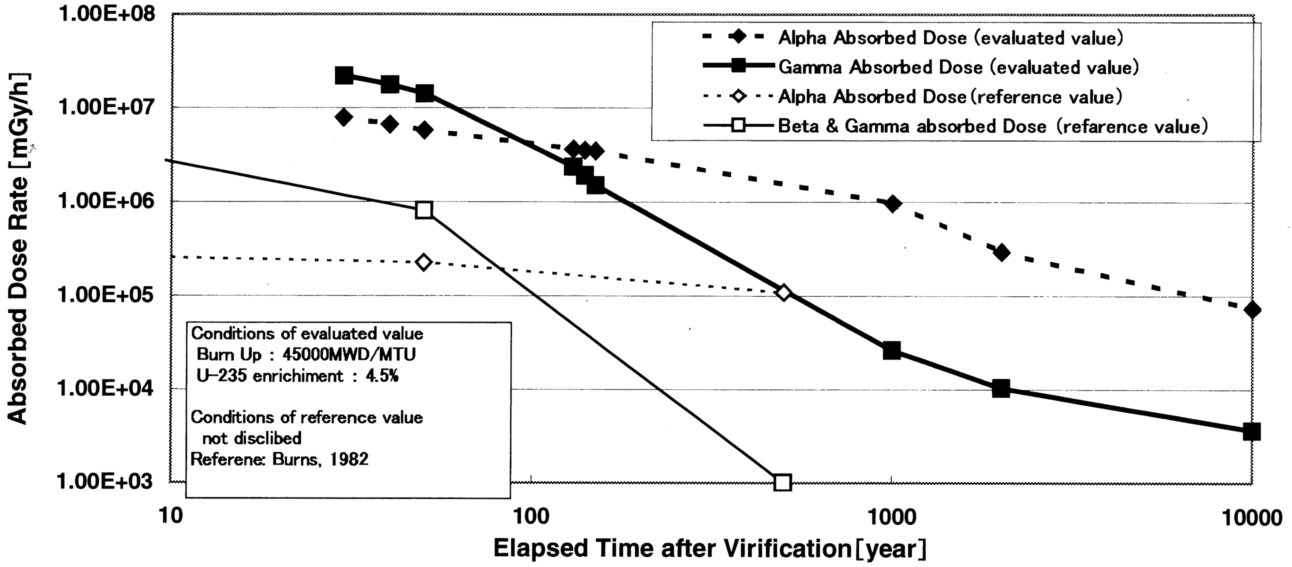

Fig. 3 Typical Radiation Absorbed Dose Rates of vitrified HLW

Table 6 Absorbed dose rate outside of Vitrified HLW [mGy/h]

Cooling Time / Vitrified: 30~100000 [Yeras]   Burn Up: 45,000 [MWD/MTU]   U-235 enrichment: 4.5 [%]
Buffer Material / Concentlation of Quartz: 0[Volume%]
Buffer Material / Water Saturation (30 years after Vitrified): 0[Volume%]
Buffer Material / Water Saturation (130 years after Vitrifed): 100[Volume%]
Corrosion product of overpack was diffused to the buffer material: 0, 30[Volume%]
Evaluation point: SIDE surface of overpack   overpack thickness: 30cm

Absorbed Dose Rate [mGy/h]	Elapsed Time from vitrified [years]				
	30	130	1000	10000	100000
Neutron - (Alpha, n) Reaction	3.244E-02	1.531E-02	3.985E-03	2.654E-04	5.634E-05
Neutron - Spontaneous Fission	3.993E-02	1.698E-03	6.308E-04	1.736E-04	3.738E-06
Neutron - TOTAL	2.329E-02	7.120E-04	1.112E-06	1.172E-06	2.607E-06
Secondary Gamma - (Alpha, n) Reaction	9.566E-02	1.772E-02	4.617E-03	4.402E-04	6.269E-05
Secondary Gamma - Spontaneous Fission	9.158E-03	6.550E-03	1.717E-03	1.136E-04	2.402E-05
Primary Gamma - Fission Product	1.199E-02	7.575E-04	2.815E-04	7.747E-05	1.668E-06
Gamma - TOTAL	2.115E-02	7.307E-03	1.998E-03	1.910E-04	2.569E-05
Absorbed Dose Rate - TOTAL	1.168E-01	2.503E-02	6.615E-03	6.312E-04	8.837E-05

product of overpack was deposited, absorbed dose rate would be increased about 65~80% compared with no corrosion products case. Because backscatter of gamma-ray was increased by Fe included in corrosion products. Therefor, if corrosion product of overpack have deposited in buffer material, absorbed dose rate should be decreased in the buffer material.

5. Embrittlement of Overpack by Neutron Irradiation

In general, neutron irradiation of carbon-steels leads to an increase in mechanical yield and fracture strength with reduced ductility, referred to as radiation hardening and embrittlement[(4)]. These effects are attributed to atomic displacement due to neutron irradiation. In addition, neutron irradiation may increase the ductile-brittle transition temperature(DBTT). The change of DBTT is rerated to neutron energy and fluence. It is possible to evaluate radiation damage, by using displacement per atom (DPA). Neutron fluence and displacement cross section are necessary to calculation DPA[(5)]. Neutron fluence in the overpack was calculated over irradiation period for the purpose of evaluating DPA.

6. Irradiation Field inside of Vitrified HLW

Alpha particle is significant to study radiolysis of grandwater invaded to vitrified HLW mentioned in section1 of chapterII. Following calculation results, gamma absorbed dose inside of vitrified HLW shared 50% in total absorbed dose rate until 50 years after vitrified. More than 100 years after vitrified, alpha absorbed dose rate become dominant in total absorbed dose rate (shown in **Fig. 3**). Because, half-life of alpha emitting actinide isotope are very long periods as pointed out by Burns[6]. It was confirmed that evaluated value were similar to reference value as described in reference (3).

7. Irradiation Field outside of Vitrified HLW

The absorbed dose rate neutron and gamma-ray, which must be taken into consideration in the analysis of radiolysis of groundwater in buffer material and host rock. Typical results for outside of vitrified HLW are shown in **Table 6**.

IV. Conclusion

Radiation can cause chemical and physical effects on HLW geological disposal system, in particular, engineered barrier system (EBS) which consists of vitrified waste, overpack container and surrounded buffer material. To assess radiation effects within the EBS for long time, it is essential to evaluate the evolution of irradiation field,quantitatively. Thus, radiation transport calculations were done to obtain dose rate, irradiation dose and absorbed dose in the irradiation field. Following three subjects obtained in this study should be significant information for assessing the radiation effects within the EBS.

(1) Radiation transport calculations were performed to evaluate the dose rate around the over-pack and vitrified HLW.

(2) Neutron fluence in the overpack was calculated over irradiation long periods for the purpose of evaluating DPA (displacement per atom).

(3) The absorbed dose of buffer material were calculated f or the purpose of evaluating the radiolysis in buffer material.

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