Hazard Evaluation on Geologic Disposal of High-Level Radioactive Waste

by

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Abstract

For a long-term safety assessment of geologic disposal of high-level radioactive waste (HLW), it is necessary to evaluate the potential hazard of radionuclides discharged into the biosphere. Therefore, the result of the safety assessement may heavily depend on this evaluation basis for the potential hazard of radionuclides. The most frequently used measure of the potential hazard is the so-called Ingestion Hazard Index based on the Maximum Permissible Concentration (MPC). Recently, however, the International Commission on Radiological Protection gave the Annual Limits of Intake for workers (ALI) . The two different bases of MPC and ALI for the potential hazard bring about considerably different results concerning the safety analysis on the geologic disposal.

We derived the explicit form of the solution for the migration of the 4-member decay chain through the one-dimensional sorbing media with dispersion. By applying the solution, it was revealed that in a limited condition of geologic formation, the most dangerous radionuclide in the geologic disposal may be altered by a change of the evaluation basis for the potential hazard. It was also pointed out that the importance of the wasteform performance may be much increased by the alteration of the evaluation basis from MPC to ALI

I. Introduction

High-Level Radioactive Wastes (HLW) generated in nuclear power plants are projected to be disposed into deep underground repositories. For a long-term safety assessment on this geologic disposal, it is necessary to predict the radiological hazard of buried HLW to mankind. Many researchers have made analyses on how much of radionuclides may be discharged into the biosphere in the future. For a safety assessment, radioactivities of discharged nuclides should be converted into their potential hazard, because 1Ci of radium-226 (^{226}Ra), for example, is much more toxic than 1Ci of tritium (^{3}H). One of the generic and convenient measures for the potential hazard through ingestion is the so-called Ingestion Hazard Index (IHI) which is defined by using the Maximum Permissible Concentration in water (MPC_w)[1] .

Table I MPC_w, ALI and F_i for some radionuclides

	Nuclide	MPC_w[a] (Ci/m^3)	ALI[b] (Bq/yr)	F_i
1	^{14}C	8×10^{-3}	9×10^{7}	0.42
2	^{79}Se	4×10^{-3}[c]	2×10^{7}	0.77
3	^{90}Sr	4×10^{-6}	1×10^{6}	−0.93
4	^{99}Tc	2×10^{-3}	1×10^{8}	−0.23
5	^{107}Pd	1×10^{-2}[c]	1×10^{9}	0.30
6	^{126}Sn	4×10^{-4}[c]	1×10^{7}	0.073
7	^{129}I	4×10^{-6}	2×10^{5}	−0.23
8	^{135}Cs	1×10^{-3}	3×10^{7}	−0.0058
9	^{137}Cs	2×10^{-4}	4×10^{6}	0.17
10	^{225}Ra	5×10^{-6}[d]	3×10^{5}	−0.31
11	^{226}Ra	1×10^{-7}	7×10^{4}	−1.37
12	^{229}Th	5×10^{-6}[c]	2×10^{4}	0.87
13	^{230}Th	2×10^{-5}	1×10^{5}	0.77
14	^{233}U	4×10^{-5}	4×10^{5}	0.47
14	^{234}U	4×10^{-5}	4×10^{5}	0.47
15	^{237}Np	3×10^{-5}	3×10^{3}	2.47
16	^{239}Pu	5×10^{-5}	2×10^{5}	0.87
17	^{241}Am	4×10^{-5}	5×10^{4}	1.40
17	^{243}Am	4×10^{-5}	5×10^{4}	1.40

(a) Ref. (1)
(b) Ref. (2)
(c) Values estimated by the authors using the parameters in Ref. (6)
(d) Ref. (5)

In 1979, based on the ICRP. Pub. 30 model[2], the International Commission on Radiological Protection (ICRP) set up an alternative limit for each radionuclide in place of MPC, which is called the Annual Limit on Intake for workers (ALI) . The ALI's are based on the biological information accumulated during these twenty years and new dose calculating models.

First, in this report, we examine the relative difference in the toxicity evaluation between ALI and MPC. Next, by using the two IHI's based on ALI and MPC, we discuss the potential toxicity of 1) HLW in storage and 2) radionuclides discharged into the biosphere after migration through geologic media. One-dimensional analysis on the migration of radionuclides in decay chains was carried out to estimate the discharge rates of radionuclides into the biosphere. The effect of wasteform performance on the long-term assessment is also discussed by using the results of the calculations.

II. Potential Hazard of HLW in Storage

(1) Difference between ALI and MPC in toxicity evaluation of radionuclides

Comparing ALI with MPC_w for each radionuclide, it may be found that one is not necessarily proportional to the other. It shows that for some nuclides the evaluation on radiological toxicity is different from each other. To quantify the difference, we introduce a factor F_i defined as follows;

$$F_i \equiv log\frac{(MPC_w)_i}{(ALI)_i/0.8} \quad , \tag{1}$$

where the suffix i denotes the nuclide i. The value of ALI is not expressed in terms of radioactivity concentration because drinking water is merely one source of ingested material, and it is difficult to identify the hysteresis of water contained in various foods. To convert the dimension into concentration (Ci/m^3), ALI is divided by the annual water intake of $0.8m^3$ for a typical adult, which is comparable with the corresponding MPC_w. The values of ALI, MPC_w and F for several nuclides are listed in Table I. The values of MPC_w are quoted from the Japanese regulations,[3] except for ^{79}Se, ^{107}Pd, ^{126}Sn, ^{225}Ra and ^{229}Th. Although the MPC_w's of these five radionuclides are not listed in Ref. (3), they are essential next to the trans-uranic elements in a long-term safety assessment of geologic disposal as pointed out by Serne & Relyea[4]. The value of MPC_w for ^{225}Ra is available in Ref. (5). For others which are not found in Ref. (3), we estimated them by using the parameters given in Ref. (6). The values of ALI for all nuclides are quoted from Ref. (2). When F_i is positive, the toxicity of the nuclide i is evaluated as more hazardous in ALI than in MPC_w. The values of F_i's for 266 radionuclides are plotted against the atomic number in Fig. 1.

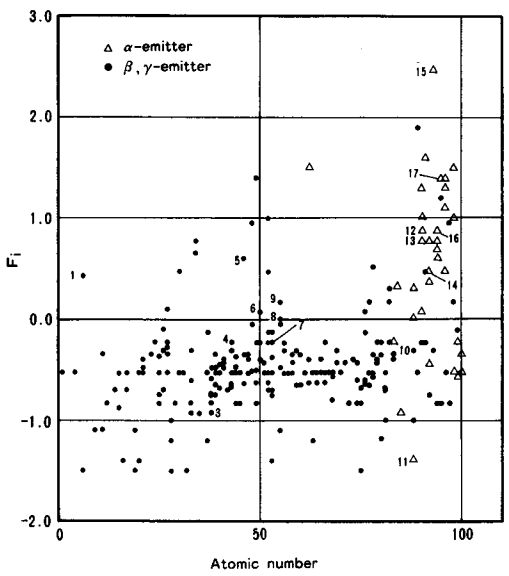

Fig. 1 Relation between the parameter F_i and atomic number. Numeral in this figure denotes the radionuclide numbered in the first column of Table I. For example, 15 in this figure denotes ^{237}Np.

As shown in the figure, the F_i's of the actinide elements of α-emitters are generally positive. It shows that their hazards are regarded as being more serious in ALI than in MPC_w. Of particular note is ^{237}Np for which F is 2.47. It tells that the hazard is evaluated approximately 300 times greater in ALI than in MPC_w. There are several other radionuclides which have high values of F; $F=40$ for ^{231}Pa, 32 for $^{249,250,251}Cf$, and 25 for $^{241,243}Am$ and $^{244,245,246,247}Cm$.

On the other hand, the values of F_i's for ^{226}Ra and some fission products (FP) such as ^{90}Sr and ^{99}Tc are negative; $F=-0.93$ for ^{90}Sr, -0.23 for ^{99}Tc and ^{129}I, and -1.37 for ^{226}Ra.

(2) Time dependence of potential hazard of HLW in storage

The MPC-based Ingestion Hazard Index of a radionuclide i, $(IHI)_{MPC,i}$ is defined as

follows;

$$(IHI)_{MPC,i} = \frac{R_i}{(MPC_W)_i/10} \quad , \tag{2}$$

Table II Inventory, Retardation Factor and Release Rate Used for Calculation

Nuclide	Half-life[a] (yr)	Inventory[b] (Bq/GWe • yr)	$1/K_i$(−) Granite[c]	Subsoil[d]	κ_i[e] (1/yr)
^{14}C	5.73×10^{3}	4.85×10^{12}	1×10^{-1}[d]	1×10^{-1}	1×10^{-6}
^{90}Sr	2.88×10	7.73×10^{16}	1×10^{-2}	1×10^{-2}	3×10^{-4}[f]
^{99}Tc	2.14×10^{5}	1.44×10^{13}	1	1	6×10^{-8}
^{129}I	1.70×10^{7}	3.77×10^{10}	1	1	1×10^{-7}[f]
^{135}Cs	3.00×10^{6}	2.88×10^{11}	1×10^{-3}	1×10^{-3}	1×10^{-4}[f]
^{137}Cs	3.02×10	1.08×10^{17}	1×10^{-3}	1×10^{-3}	1×10^{-4}[f]
^{225}Ra	4.05×10^{-2}	0	1×10^{-3}	2×10^{-3}	3×10^{-6}[f]
^{226}Ra	1.60×10^{3}	0	1×10^{-3}	2×10^{-3}	3×10^{-6}[f]
^{229}Th	7.30×10^{3}	0	2×10^{-4}	2×10^{-5}	3×10^{-6}[f]
^{230}Th	8.00×10^{4}	0	2×10^{-4}	2×10^{-5}	3×10^{-6}[f]
^{233}U	1.59×10^{5}	0	2×10^{-2}	7×10^{-5}	1×10^{-8}
^{234}U	2.45×10^{5}	7.18×10^{9}	2×10^{-2}	7×10^{-5}	1×10^{-8}
^{237}Np	2.14×10^{6}	5.33×10^{11}	4×10^{-3}	1×10^{-2}	6×10^{-8}
^{239}Pu	2.41×10^{4}	3.26×10^{12}	9×10^{-4}	1×10^{-4}	1×10^{-6}
^{241}Am	4.33×10^{2}	1.68×10^{14}	1×10^{-5}	1×10^{-4}	3×10^{-8}
^{243}Am	7.37×10^{3}	1.76×10^{13}	1×10^{-5}	1×10^{-4}	3×10^{-8}

(a) Ref. (8)
(b) Uranium-fueled 1 GWe PWR, 150 days after discharge. Ref. (5)
(c) Ref. (9)
(d) Ref. (10)
(e) Referred to Ref. (11)
(f) Assumed values

where R denotes the radioactivity. The MPC_W which must originally be applied to workers is divided by ten to obtain a concentration limit for the general public. Then the $(IHI)_{MPC,i}$ expresses a relative index in terms of water volume which is required to dilute R_i to a concentration limit for the public.

Now we introduce another IHI defined as follows;

$$(IHI)_{ALI,i} \equiv \frac{R_i}{(ALI)_i/0.8/10} \quad , \tag{3}$$

in which the factor 0.8 is the same as that in Eq. (1).

The time-dependence of radioactivity of nuclide i in a decay chain can be

expressed as follow;

$$R_i = \lambda_i S_1^0 \sum_{j=1}^{i} B_{i,j}\, exp\ (-\lambda_j \tau) \tag{4}$$

In the above equation, $B_{i,j}$ is the Bateman Coefficient[7] denoted by

$$B_{i,j} = \sum_{m=1}^{j} \frac{(S_m^0/S_1^0) \prod_{r=m}^{i} (\lambda_r/\lambda_i)}{\prod_{\substack{t=m \\ t \neq j}}^{i} (\lambda_t - \lambda_j)} \quad , \quad B_{1,1} = 1, \tag{5}$$

where λ_i is a decay constant and S_i^0 is the number of nuclide i at 150 days ($\tau=0$) after the discharge of spent fuel from a power plant. The inventory of radionuclides in spent fuel listed in Table II is based on radioactivities produced in a 1GWe nuclear power plant for 1yr. Volatile elements such as carbon and iodine are included in Table II since they also have the possibility of being subjected to a geologic disposal.

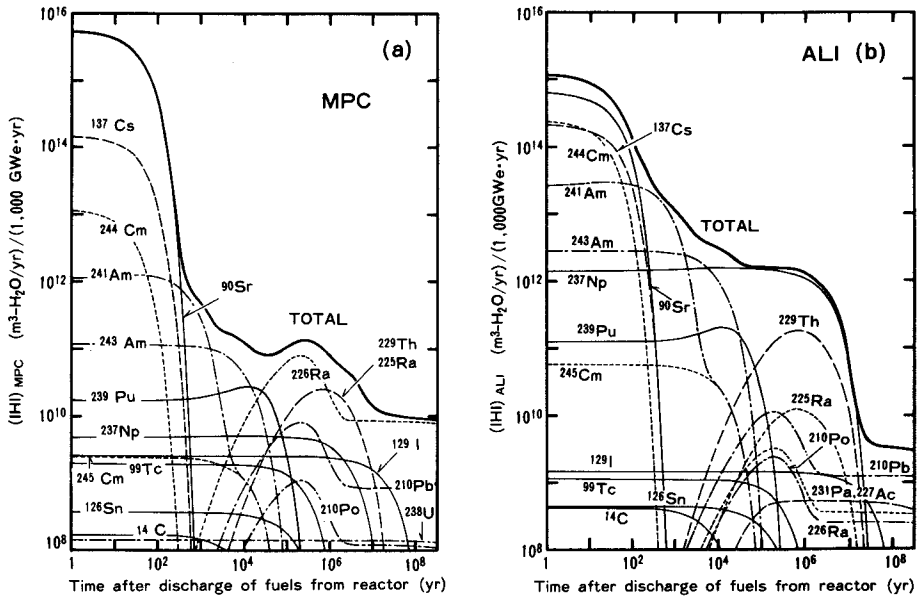

Fig. 2 Ingestion Hazard Indices of HLW as a function of time. (a); based on MPC, (b); based on ALI.

By applying Eqs. (2)–(5), it is possible to calculate $(IHI)_{MPC}$ and $(IHI)_{ALI}$ for the waste generated by the operation of nuclear plants of 1,000GWe· yr as a function of time. The results are shown in Fig. 2 (a) and (b), in which the hazard index for various individual radioactive isotopes is plotted as well as the total for all combined. Cohen has obtained a similar result by applying the cancer risk[8]. In $(IHI)_{MPC}$, ^{90}Sr dominates by 96% of the total or more at the early stage of storage, while the contributions of ^{137}Cs and ^{244}Cm are not negligible as well as ^{90}Sr in $(IHI)_{ALI}$. It results from the fact that the hazard of ^{90}Sr is 0.1 times smaller and that of ^{244}Cm is 25 times

greater in ALI than in MPC, as seen in F_i in Table I.

During the first 1,000 years the total $(IHI)_{MPC}$ is reduced to a factor 10^{-4} of the initial inventory ($\tau=0$). It is mainly due to the decay of ^{90}Sr and ^{137}Cs. As for the total $(IHI)_{ALI}$ however, one cannot expect such a rapid reduction during the period. It decreases to about 10^{-2} times of the initial. It may take 10^7 yr for the decrease of the total $(IHI)_{ALI}$ to 10^{-4} times.

As seen in Fig. 2 (a) and (b), during the term around 1,000yr to 10,000yr, the isotopes of americium dominate both of the IHI's. However, the term from 10^4 yr to 10^6 yr is dominated by ^{226}Ra in $(IHI)_{MPC}$ and by ^{237}Np in $(IHI)_{ALI}$, respectively. It should be noticed that these two dominants have different origins, that is, ^{237}Np is originally contained in the spent fuels, and ^{226}Ra is generated by the decay of ^{234}U after the discharge of spent fuels.

On the durability of waste packages buried in a deep geologic formation, the criteria "No release of radionuclides from the waste package for 1,000 years after the repository is sealed"[13] has been discussed. When one refers to the time-dependence of $(IHI)_{MPC}$ shown in Fig. 2 (a), the effectiveness of the criterion of '1,000 years' enclosure can be verified. As shown in Fig. 2 (b), however, the term of the criterion seems not to be so effective for the decrease of the potential hazard expressed by $(IHI)_{ALI}$. It should be noticed that the decay scheme of the potential hazard of HLW depends much on the choice of the hazard index.

Ⅲ. Potential Hazard due to Discharge of Radionuclides into the Biosphere

We cannot expect a permanent soundness of the repository system. It must be considered that groundwater will penetrate through the engineering barriers constructed by backfill materials, buffer materials, canister and others, and attack the wasteform to dissolve the radionuclides. The dissolved radionuclides may migrate along with the groundwater through geologic formations and may be finally discharged into the biosphere. Therefore, the prediction of the discharge rate of radionuclides into the biosphere is of primary importance in the safety assessment of geologic disposal.

Although the migration behavior of radionuclides through geologic media depends on various phenomena, an extremely simplified model is used for the following analysis. Assuming a one-dimensional column for the groundwater path, the fundamental differential equation on the migration of radionuclide i in a decay chain of A $(i=1) \rightarrow B$ $(i=2) \rightarrow C$ $(i=3) \rightarrow \cdots$ can be expressed as follows;

$$K_i\frac{\partial N_i}{\partial t} = D\frac{\partial^2 N_i}{\partial z^2} - V\frac{\partial N_i}{\partial z} - \lambda_i K_i N_i + \lambda_{i-1} K_{i-1} N_{i-1} \tag{6}$$

$$\lambda_0 \equiv 0,\ i = 1,2,\cdots,\ 0 \leqq z,\ 0 \leqq t,$$

where

N_i: radionuclide concentration dissolved in groundwater (nuclides/m^3)

λ_i: decay constant (1/yr)

K_i: retardation factor (−)

D: dispersion coefficient (m^2/yr)

V: groundwater velocity (m/yr)

t : time after the start of release from repository (yr)

z : path length from repository (m)

The initial and boundary conditions are given by

Initial condition:

$$N_i\ (z,\ 0) = 0 \tag{7}$$

Boundary condition:

$$N_i\ (0,\ t) = (\kappa_i M_1^0/Q) \sum_{j=1}^{i} B'_{i,j} exp\ (-\Lambda_j t) \tag{8}$$

$$N_i\ (\infty, t) = 0 \tag{9}$$

The Preferential Release Model[14] is applied in the boundary condition Eq. (8), in which the amount of release per unit time from the repository is proportional to that remaining in the wasteform. The parameter κ_i is called the release rate coefficient for the Preferential Release Model. In Eq. (8), Q is the annual volumetric flow rate of groundwater, and Λ_i and $B'_{i,j}$ are respectively defined as

$$\Lambda_i \equiv \lambda_i + \kappa_i \tag{10}$$

$$B'_{i,j} \equiv \sum_{m=1}^{j} \frac{(M_m^0/M_1^0) \prod_{r=m}^{i} (\lambda_r/\lambda_i)}{\prod_{\substack{l=m \\ l \neq j}}^{i} (\Lambda_l - \Lambda_j)}\ ,B'_{1,1} = 1\ , \tag{11}$$

where M_m^0 is the amount of the nuclide m in the wasteform at the beginning of release into geologic media ($t=0$). In the Appendix, the solution of Eq. (6) for the 4-member decay-chain under the conditions of Eqs. (7)-(9) is shown in an explicit form.

The inventory is normalized into that corresponding to the operation of nuclear plants of 1,000GWe·yr. Radionuclides are assumed to be kept in the wasteform for 1,000 years, that is to say, the release of radionuclides from the wasteform to geologic medium begins 1,000 years after the discharge of spent fuel from a reactor. In order to simplify the situation, it is also assumed that the radionuclides are directly released into a host rock of granite. The retardation factors for granite used in the calculations are listed in Table II, which have been estimated by KBS[9]. The groundwater velocity and dispersion coefficient are set as 1m/yr and 100m^2/yr, respectively.

The release rate coefficient κ for each element in Table II is assumed with reference to the solubility- limited dissolution theory given by Chambré et al.[11]

Now, let us calculate, as an example, the discharge rates of radionuclides into the biosphere at a location of 1,000m apart from the repository. The results are shown in Fig. 3(a) and (b), in which hazard indices corresponding to the annual discharge rates of radionuclides are presented for the individual radionuclide as well as the total for all. One may notice that an extremely high peak of ^{237}Np appears in Fig. 3 (b), while in Fig. 3(a) several radionclides contribute to the maximum potential hazard in a comparable extent. It should also be noticed that those important radionuclides expected through the calculation of migration are not always the same as those regarded as predominant nuclides in the discussion on Fig. 2(a) and (b). Except for the volatile nuclides (^{129}I and ^{14}C), the most predominant nuclides in Fig. 3(b) are those in the decay-chain of ^{237}Np→^{233}U→^{229}Th→^{225}Ra. Therefore, the partitioning of ^{237}Np from HLW in the reprocessing process may be expected to considerably reduce the risk due to HLW.

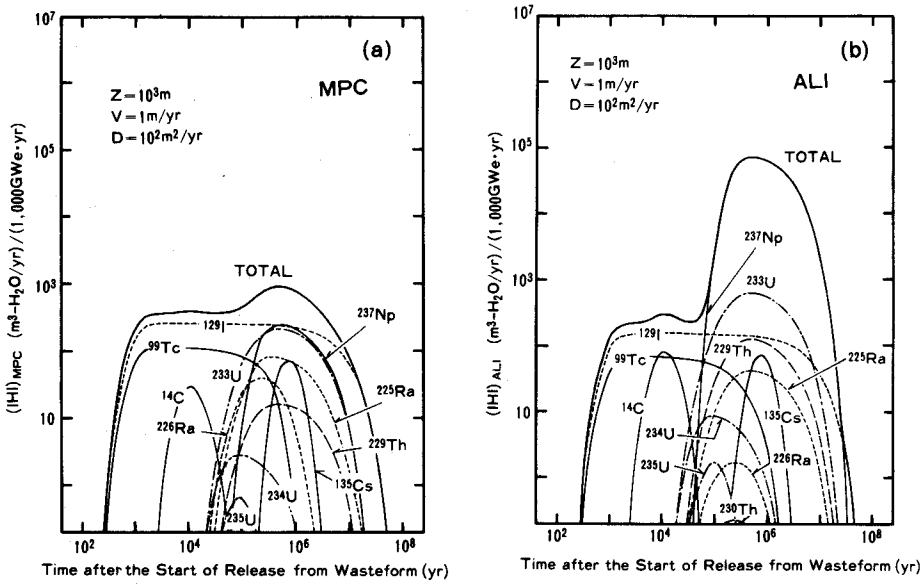

Fig. 3 Annual discharge rates of radionuclides at a location of 1,000 m from wasteforms. Host rock; granite. a); based on MPC, b); based on ALI.

IV. Effect of Wasteform Performance on the Potential Hazard of Radionuclides in the Biosphere

In some previous discussions[10,15] based on $(IHI)_{MPC}$ on the long-term safety assessment of geologic disposal, ^{226}Ra has been recognized as the most critical nuclide. Especially when the retardation factors for subsoil[10] are used for the calculation, the

so-called reconcentration phenomenon of ^{226}Ra occurs. It may bring about the possibility of a high discharge rate of ^{226}Ra at a location fairly far from the repository at around 2×10^5 years after the burification of HLW.

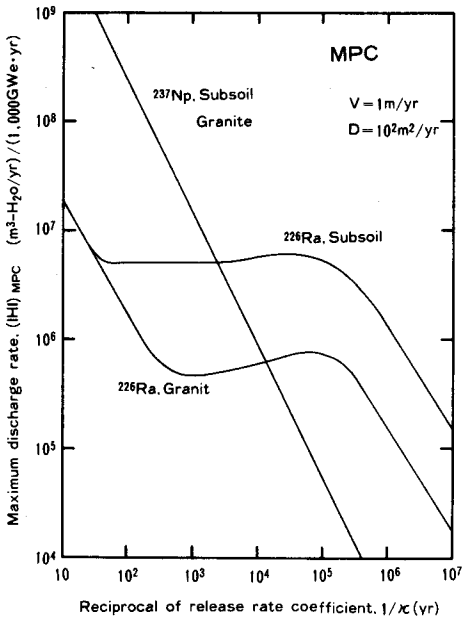

Fig. 4 The maximum discharge rates of ^{226}Ra and ^{237}Np as a function of the reciprocal of release rate coefficient κ. Retardation factors given in Ref. (10) were used for subsoil. Host rock; granite and subsoil.

By applying the solution given in the Appendix, we analyzed the maximum concentration of ^{226}Ra and ^{237}Np in granite and subsoil in the whole range of time t and distance from wasteform z. The results are presented in Fig. 4, where the concentration is expressed as the Ingestion Hazard Index $(IHI)_{MPC}$ corresponding to the annual discharge rate $(Q\lambda_i N_i)$. The congruent release is assumed in these calculations, where the release rate coefficient for every nuclide is same as each other $(\kappa_1=\kappa_2=\cdots=\kappa)$. As seen from this result, ^{226}Ra is a very important radionuclide in the safety assessment.

It is interesting to note that a broad plateu is seen in the curve of ^{226}Ra for subsoil. It means that the wasteform performance has little effect on the maximum concentration of ^{226}Ra insofar as the release rate coefficient for the wasteform cannot be reduced to less than 10^{-5} (1/yr).

On the other hand, the maximum concentration of ^{237}Np appears at the surface of the wasteform at the beginning of the release, because most of ^{237}Np is present in HLW itself and is not produced in the geologic medium. Therefore, the maximum concentration is strictly proportional to the reciprocal of the leachability. When ALI is applied in the calculation in place of MPC, the potential hazard of ^{237}Np in the result is much higher than that of ^{226}Ra in the whole range of the release rate coefficient in Fig. 4. So far as ALI will be used in the safety assessment, the improvement of the wasteform performance must be directly effective to reduce the risk of the disposal of HLW.

V. Conclusion

The safety analysis on geologic disposal of HLW is affected by the alteration of the evaluation basis on radiological toxicity.

Most of actinide α-emitters, especially ^{237}Np, are regarded to be more serious in ALI than in MPC_w. On the other hand the toxicities of ^{226}Ra and β-emitters such as ^{90}Sr and ^{99}Tc are evaluated lower in ALI than in MPC_w. These differences result in that $(IHI)_{ALI}$ of HLW decreases to a factor 10^{-2} during the first 1,000 years, while $(IHI)_{MPC}$ decreases to a factor 10^{-4} during the same term. The difference in expectation on the decrease of toxicity of HLW may be important for the required life-time of the engineering barrier constructed in the repository.

By the analysis of radionuclide migration through geologic medium, it was found that the maximum discharge rate of ^{226}Ra has a tendency to be independent of wasteform leachability when the release rate coefficient is not extremely small. As for ^{237}Np , in contrast to ^{226}Ra, the maximum discharge rate is strictly proportional to the reciprocal of the release rate coefficient. Depending on the geologic condition assumed, ^{226}Ra may be a very critical nuclide if MPC is used in the safety analysis. In such cases, the decrease of leachability will not contribute so much to the risk reduction. When the analysis is based on ALI in place of MPC, however, ^{237}Np will be extremely more important than the others. So far as based on ALI, the improvement of the wasteform performance is expected to be quite effective for reducing the risk due to the geologic disposal of HLW.

The calculations were performed by the computer at The Data Processing Center of Kyoto University.

Appendix

In this Appendix, the solution for the 4-member decay chain of Eq. (6) is presented. This solution can be applied to the decay chain whose members have different retardation factors for each and all.

$$N_i\ (z,t) = N_i^0 \sum_{j=1}^{i} B_{i,j} E\ (i,\ j,\ 0)$$

$$+ N_{i-1}^0 h_{i,\,i-1} \sum_{j=1}^{i-1} B_{i-1,j}\ (i,\ i-1,\ j,\ 0)$$

$$\times \{E\ (i-1,\ j,\ 0)\ - E\ (i,\ j,\ 0)\ + E\ (i,\ i,\ i-1)\ - E\ (i-1,\ i,\ i-1)\ \}$$

$$+ N_{i-2}^0\, h_{i-1,\,i-2}\, h_{i,\,i-2} \sum_{j=1}^{i-2} B_{i-2,j}$$

$$\times\begin{bmatrix} d(i-1,i-2,j,0)\ d(i,i-2,j,0)\{E(i-1,j,0)-E(i,j,0)\} \\ +d(j,0,i-1,i-2)d(i,i-2,i-1,i-2)\{E(i-2,i-1,i-2)-E(i,i-1,i-2)\} \\ +d(j,0,i,i-2)d(i-1,i-2,i,i-2)\{E(i-2,i,i-2)-E(i,i,i-2)\} \end{bmatrix}$$

$$+N^0_{i-2}\,h_{i-1,i-2}\,h_{i,i-1}\sum_{j=1}^{i-2}B'_{i-2,j}$$

$$\times\begin{bmatrix} d(i-1,i-2,j,0)d(i,i-1,j,0)\{E(i,j,0)-E(i-1,j,0)\} \\ +d(j,0,i-1,i-2)d(i,i-1,i-1,i-2)\{E(i,i-1,i-2)-E(i-1,i-1,i-2)\} \\ +d(j,0,i,i-1)d(i-1,i-2,i,i-1)\{E(i,i,i-1)-E(i-1,i,i-1)\} \end{bmatrix}$$

$$+N^0_{i-3}h_{2,1}h_{3,1}h_{4,1}$$

$$\times\begin{bmatrix} d(2,1,1,0)d(3,1,1,0)d(4,1,1,0)\{E(1,1,0)-E(4,1,0)\} \\ +d(1,0,2,1)d(3,1,2,1)d(4,1,2,1)\{E(1,2,1)-E(4,2,1)\} \\ +d(1,0,3,1)d(2,1,3,1)d(4,1,3,1)\{E(1,3,1)-E(4,3,1)\} \\ +d(1,0,4,1)d(2,1,4,1)d(3,1,4,1)\{E(1,4,1,)-E(4,4,1)\} \end{bmatrix}$$

$$+\mathrm{N}^0_{i-3}\mathrm{h}_{2,1}\mathrm{h}_{3,1}\mathrm{h}_{4,3}$$

$$\times\begin{bmatrix} d(2,1,1,0)d(3,1,1,0)d(4,3,1,0)\{E(4,1,0)-E(3,1,0)\} \\ +d(1,0,2,1)d(3,1,2,1)d(4,3,2,1)\{E(4,2,1)-E(3,2,1)\} \\ +d(1,0,3,1)d(2,1,3,1)d(4,3,3,1)\{E(4,3,1)-E(3,3,1)\} \\ +d(1,0,4,3)d(2,1,4,3)d(3,1,4,3)[E(4,4,3)-E(3,4,3)\} \end{bmatrix}$$

$$+N^0_{i-3}h_{2,1}h_{3,2}h_{4,2}$$

$$\times\begin{bmatrix} d(2,1,1,0)d(3,2,1,0)d(4,2,1,0)\{E(4,1,0)-E(2,1,0)\} \\ +d(1,0,2,1)d(3,2,1,2)d(4,2,1,2)\{E(4,2,1)-E(2,2,1)\} \\ +d(1,0,3,2)d(2,1,3,2)d(4,2,3,2)\{E(4,3,2)-E(2,3,2)\} \\ +d(1,0,4,2)d(2,1,4,2)d(3,2,4,2)\{E(4,4,2)-E(2,4,2)\} \end{bmatrix}$$

$$+N^0_{i-3}h_{2,1}h_{3,2}h_{4,3}$$

$$\times\begin{bmatrix} d(2,1,1,0)d(3,2,1.0)d(4,3,1,0)\{E(3,1,0)-E(4,1,0)\} \\ +d(1,0,2,1)d(3,2,2,1)d(4,3,2,1)\{E(3,2,1)-E(4,2,1)\} \\ +d(1,0,3,2)d(2,1,3,2)d(4,3,3,2)\{E(3,3,2)-E(4,3,2)\} \\ +d(1,0,4,3)d(2,1,4,3)d(3,2,4,3)\{E(3,4,3)-E(4,4,3)\} \end{bmatrix}$$

, where

$$N^0_i \equiv \begin{cases}\kappa_i \mathrm{M}^0_1/\mathrm{Q} & \mathrm{i}\geqq 1\\ 0 & \mathrm{i}\leqq 0\end{cases}$$

$$h_{i,j} \equiv \frac{\lambda_{i-1}/v_{i-1}}{1/v_i-1/v_i}$$

$$v_i \equiv V/K_i$$

$$d(i,\ j,\ k,\ r) \equiv (\beta_{i,j}-\beta_{k,r})^{-1}$$

$$\beta_{i,j} \equiv \frac{\lambda_j v_i - \lambda_i v_j}{v_i - v_j}$$

$$\beta_{i,\,o} \equiv \Lambda_i$$

The function $E(i,j,k)$ is as follows:

$$E(i,j,k) = \frac{2}{\sqrt{\pi}} exp\ (z/2\alpha - \beta_{j,k}t)$$
$$\times \int_{a_i z/2\sqrt{t}}^{\infty} exp\{-[\xi^2 + (b_i - \beta_{j,k})(a_i z)^2/4\xi^2]\}\, d\xi, \qquad (A-2)$$

where

$$a_i \equiv 1/\sqrt{\alpha v_i} = \sqrt{K_i/D}$$
$$b_i \equiv \lambda_i + v_i/4\alpha$$
$$\alpha \equiv D/V$$

When the inequality

$$b_i - \beta_{j,k} \equiv \gamma_{ijk} \geqq 0$$

is allowed, Eq. (A-2) can be transformed to yield

$$E(i,\ j,\ k) = \frac{1}{2} exp\ (z/2\alpha - \beta_{j,k}t)$$
$$\times [exp(a_i z\sqrt{\gamma_{ijk}})\ erfc\ (a_i z/2\sqrt{t} + \sqrt{\gamma_{ijk}t})$$
$$+ exp\ (-a_i z\sqrt{\gamma_{ijk}})\, erfc\ (a_i z/2\sqrt{t} - \sqrt{\gamma_{ijk}t})],$$

where

$$erfc\ (x) = \frac{2}{\sqrt{\pi}} \int_x^{\infty} e^{-u^2} du$$

If $\gamma_{ijk} < 0$, numerical integration must be carried out for Eq. (A-2).

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