Uncertainty and Sensitivity Analysis for Long-Term Performance of Sand-Bentonite Buffer Material[*]-10077

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ABSTRACT

In the geological disposal of high-level radioactive waste, long-term alteration of buffer material is likely to occur during the period of elevated temperatures and/or under highly alkaline environments induced by cementitious materials. Monte Carlo-based uncertainty and sensitivity analysis was carried out in order to identify important scenarios and parameters with respect to the long-term alteration of buffer material. One of the possible scenarios is the loss of impermeability and occurrence of advective groundwater flow in the altered buffer material, and resulting increased glass dissolution rate. This scenario allows a conspicuous increase in the maximum release rates of Cs-135 and Se-79 from the buffer material to the surrounding host rock compared with those in the normal scenario. Three time-dependent parameters correlated with the alteration, i.e. pore velocity, effective diffusion coefficient and glass dissolution rate were sensitive to the maximum release rate of these radionuclides. Decreased distribution coefficients of Cs and Se caused by the alteration, on the other hand, were insensitive parameters on the maximum release rates of these radionuclides.

INTRODUCTION

Nuclear Waste Management Organization of Japan as an implementer has been accepting open solicitation for candidate sites of final disposal of high-level radioactive waste (HLW) [1]. Nuclear Safety Research Center (NSRC) of Japan Atomic Energy Agency (JAEA) has carried out regulatory research in order to provide technical support for safety review of the license application for the final disposal site. Our regulatory research is to develop tools (scenarios, models and parameters) to assess the long-term performance of a geological disposal system.

In safety assessment for a geological disposal of HLW, it is of consequence to estimate the uncertainties due to the long-term frame associated with long-lived radionuclides and the expanded geological environment. Research activities of our group in NSRC have been focused on the long-term performance of the engineered barrier system. One of the research objects is the long-term chemical alteration of sand-bentonite mixture which is as a candidate of buffer material in Japan. The alteration of the buffer material may progress during the period of elevated temperatures and/or under highly alkaline environments induced by cementitious materials in the repository. The long-term alteration of buffer material may have detrimental effects on its physical and/or chemical properties. Variation of the hydraulic conductivity caused by the alteration is considered to affect the mass transport from glass to the host rock. Nakayama et al. [2] and Yamaguchi et al. [3] in our research group have modeled the hydraulic conductivity of compacted sand–bentonite mixtures through their experiments on montmorillonite

dissolution under alkaline conditions. The model predicts the long-term performance of the buffer material under geological disposal conditions.

This study carried out an uncertainty and sensitivity analysis with the objective of identifying important scenarios and parameters with respect to long-term alteration of the buffer material. The Monte Carlo technique was employed in the analysis.

SCENARIO DESCRIPTION

Geological disposal system assumed for this study is that designed in the H12 Project report [4] of Japan Nuclear Cycle Development Institute. The repository is constructed in a deep and stable granitic bedrock at a depth of 1,000 m. The rock is described in terms of two major hydraulic units; fractured zone and rock mass. It is reasonable to assume that the repository is constructed in a stable rock mass having enough distances from fractured zones, in order to avoid the occurrence of a short path of groundwater from the repository to the biosphere.

The engineered barrier system is composed of borosilicate vitrified waste form, carbon steel overpack and bentonitic buffer material (a mixture of 70 wt% of bentonite and 30 wt% of sand). Cementitious materials are made use of for the plugs, grouting and support of tunnel.

In this analysis, it is considered that the alteration scenario of the buffer material is caused by highly alkaline groundwater from the cementitious materials. The dissolution of montmorillonite and mineralogical transformations may lead to undesirable properties of the buffer material, e.g., higher hydraulic conductivity and lower swelling pressure.

Different four analytical cases are set as shown in Table 1. The alteration scenario includes three analytical cases: diffusive transport in the altered buffer (referred to as "Case B1"), advective transport (referred to as "Case B2") and increased dissolution rate of the glass matrix correlated with the advective transport (referred to as "Case B3"). In order to compare with the alteration scenario, the case of normal scenario (referred to as "Case A") is also considered in this analysis under the assumption that no advertent events are anticipated and radionuclides are transported in the intact geological disposal system.

Table 1 Description of the normal scenario and three alteration scenarios targeted in this analysis

Scenario	Description	Case
Normal scenario	No advertent events are anticipated and radionuclides are transported in the intact geological disposal system.	Case A
Alteration Scenario	The alteration causes an increase in the diffusion coefficient of radionuclides in the altered buffer material	Case B1
	Diffusive transport of radionuclides are dominant over advective transport.	Case B2
	Advection of groundwater decreases the concentration of silica in the porewater in the buffer material, resulting an increased glass dissolution.	Case B3

In the scenario of Case B1, it is assumed that the alteration causes an increase in the diffusion coefficient of radionuclides in the altered buffer. In CaseB2, transport of radionuclides in the buffer material changes from diffusion limited to advection limited as the alteration progresses. In the case limited by the diffusive transport in the buffer material, the concentration of dissolved silica in the

vicinity of the glass approaches the saturation level, and the glass dissolution rate comes to be lower, owing to the protective effect of an altered layer formed on the glass surface, rather than that controlled by the first-order dissolution rate law [5]. In CaseB3, it is supposed that the glass dissolution rate becomes to be higher, with the decreased concentration of dissolved silica.

It has not been understood whether altered bentonite enhances or depresses its sorbability of radionuclides. Depressed sorption was assumed in this alteration scenario for conservative evaluation.

CONCEPTUAL AND MATHEMATICL MODELS

Takeda et al. have developed the probabilistic safety assessment code system GSRWPSA (Generic Safety assessment code for geologic disposal of Radioactive Waste Probabilistic Safety Assessment) [6], to quantify the uncertainties in parameters and conceptual models based on the Monte Carlo calculation. We apply a compartment model of 1-D radionuclide transport in the engineered barrier system in GSRWPSA. It is assumed that the overpack is designed to have at least a minimum lifetime of 1,000y [4]. After the overpack failure, radionuclides are released from the vitrified waste form as a result of glass matrix dissolution. Radionuclides are transported in the overpack and buffer material by diffusion and/or advection depending on the progress of the long-term alteration of the buffer material. Retardation and precipitation/dissolution determined by the elemental solubility were taken into account. The sorption onto corrosion products of overpack is disregarded for conservativeness. The sorption onto corrosion products of overpack is conservatively disregarded.

Under the assumption of the congruent release with glass dissolution, the release rate of radionuclides from the vitrified waste form can be expressed as follows:

$$J_g^i(t) = \frac{v_g(t) \cdot S_g}{\rho_g V_g(t) \cdot (1 - \varepsilon_g)} A_g^i(t)$$
(Eq.1)

, where:

$J_g^i(t)$: Release rate from vitrified waste form of radionuclide i [Bq/y],

$v_g(t)$: Glass dissolution rate [kg/m^2/y],

S_g : Surface area related to dissolution of the vitrified waste form [m^2],

$A_g^i(t)$: Inventory of radionuclide i in the vitrified waste form [Bq],

ρ_g : Density of the vitrified waste form [kg/m^3],

ε_g : Porosity of the vitrified waste form [-], and

$V_g(t)$: Volume of the vitrified waste [m3].

The time-dependent inventory $A_g^i(t)$ in the vitrified waste form is given by the following equation in consideration of radioactive decay chain:

$$\frac{dA_g^i(t)}{dt} = -J_g^i(t) - \lambda^i A_g^i(t) + \lambda^i A_g^{i-1}(t)$$
(Eq.2)

, where: λ^i : Decay constant of radionuclide i [1/y].

The decreasing volume of the vitrified waste form with dissolution can be described by

$$V_g(t) = V_g(0) - \frac{S_g}{(1-\varepsilon_g)\rho_g} \int_0^t v_g(\tau)d\tau \qquad (Eq.3)$$

, where: $V_g(0)$: Initial volume of the vitrified waste [m³].

The mass balance equation in the overpack and the buffer material, "compartment", is given by Eq.4:

$$\frac{dA_k^i}{dt} = -J_{k,k+1}^i + J_{k-1,k}^i - \lambda^i A_k^i + \lambda^{\ i} A_k^{i-1} \qquad (Eq.4)$$

, where the release rate of radionuclide i from compartment k to $k+1$, $J_{k,k+1}^i(t)$ is expressed by the summation of contributions of diffusive transport and advective transport (see Eq.5).

$$J_{k,k+1}^i(t) = S_k u_k(t)\varepsilon_k C_k^i(t) - D_k^i(t)S_k \left[\frac{\Delta C_{k,k+1}^i(t)}{\Delta x} \right] \qquad (Eq.5)$$

Where:

S_k : Release area of the compartment k [m²],

$u_k(t)$: Pore velocity in the compartment k [m/y],

ε_k : Porosity of the compartment k [-],

$C_k^i(t)$: Concentration of radionuclide i in the compartment k [Bq/m³],

$D_k^i(t)$: Effective diffusion coefficient of radionuclide i in the compartment k [m²/y],

Δx : Distance of the adjacent compartments [m], and

$\Delta C_{k,k+1}^i(t)$: Difference in the concentration between the adjacent compartments [Bq/m³].

The relation between the concentration of radionuclide i, $C_k^i(t)$, and the inventory in the compartment k, $A_k^i(t)$, is given by the Eq.6:

$$C_k^i(t) = \frac{A_k^i(t)}{V_k \varepsilon_k R_k^i(t)} . \qquad (Eq.6)$$

Where:

V_k : Volume of the compartment k [m³], and

R_k^i : Retardation factor for radionuclide i [-].

The retardation factor can be written as follows,

$$R_k^i(t) = 1 + \frac{1-\varepsilon_k}{\varepsilon_k} \rho_k Kd_k^i(t) \qquad (Eq.7)$$

, where,

ρ_k : Density of the compartment k [m²], and

$Kd_k^i(t)$: Distribution coefficient of radionuclide i for the compartment k [m³/kg].

The time dependence of glass dissolution rate, diffusion coefficient, velocity and distribution coefficient in the buffer material are considered in the analysis for the alteration scenario. Those parameters are expressed as the step functions of time in GSRWPSA. It is assumed that there is no spatial variation of the parameters in the buffer material.

We investigate the relationship between the maximum release rate of radionuclides discharged from the engineered barrier system and the time-dependent parameters, through the uncertainty and sensitivity analysis.

ESTIMATION OF PARAMETER UNCERTAINTY

We carried out the preliminary uncertainty analysis for hypothetical geological disposal system of HLW [7]. The previous analysis indicates that the important radionuclides on radiological effect are identified as Se-79, Cs-135, progeny radionuclides of Np-237 and U-238. In this paper, we select Se-79 and Cs-135 for the analysis of the alteration scenario. The parameters on the design of the engineered barrier system are referred from the H12 Project report [4]. The uncertainties on five kinds of the parameters, i.e. solubility, glass dissolution rate, effective diffusion coefficient, distribution coefficient and pore velocity in the buffer material are considered in this analysis.

(1) Solubility

The solubility for Se is estimated using the probabilistic analysis code of uncertainty of solubility limit for radioactive elements in a geological disposal (PASOL), developed by Takeda et al. [8]. The PASOL computer code is on the basis of Latin Hypercube Sampling (LHS) code [9] and geochemical code of EQ3/6 [10]. The code estimates the uncertainty of solubility limit associated with the uncertainties of both groundwater chemistry and thermodynamic data. The chemical composition of groundwater is taken from the calculated bentonite porewater in the H12 Project report [4]. The uncertainties of thermodynamic data of Se are basically referred from the uncertainty estimates that are represented in the NEA TDB [11]. The solid phase of Se solubility limiting is assumed to be Se(cr). The result of solubility calculation indicates that the uncertainty on solubility of Se(cr) is governed by the error of the equilibrium constant for the reaction between Se(cr) and HSe⁻ and estimated to be in the range of 1E-7 to 6E-7 mol/l corresponding to the 95% confidence intervals. This estimated variation of Se solubility is used for the analysis of Se-79 transport in the engineered barrier system. Since Cs is a highly soluble element, the solubility limit for Cs is not considered.

(2) Glass Dissolution Rate

It is assumed that the alteration and dissolution of glass in the engineered barrier system proceeds by a mechanism neglecting the effects of the chemical affinity for dissolution reaction of glass matrix. We estimate the uncertainty of the long-term glass dissolution rate represented by a constant dissolution rate, which has been used for the safety assessment in Japan. In order to estimate the parameter uncertainty of the glass dissolution rate, we carried out the statistical analysis of the release rate data of boron for the hypothetical condition of disposal system, based on the relation between published data of glass dissolution and those experimental conditions. The conditions for data selection are a type of glass, temperature, pH, ionic strength, and saturation condition of dissolved silica. As a result of the constant

dissolution rate, the uncertainty of the glass dissolution rate is estimated to be a variation with about 3 orders and logarithmic mean of 0.004 $g/m^2/day$. This variation of glass dissolution rate is used for the analytical case of the normal scenario.

Under the assumption of increased dissolution rate of the glass matrix correlated with the advective transport in the alteration scenario, it is supposed that the maximum of the dissolution rate is conservatively equivalent to level of the first-order dissolution rate. The range of time-dependant glass dissolution rate is considered as from 0.004 to 1.0 $g/m^2/day$. Fig.1 shows the relation between the increased dissolution rate and time. In the analysis for the alteration scenario, it is assumed that the increase in glass dissolution rate (=0.004 $g/m^2/day$) starts from 1,000 y after the disposal (t=0) as the alteration progresses and the end time for increased dissolution rate is in the variation of 2,000 to 10,000 y. The relation between the glass dissolution rate and time from the start to the end time is assumed to be expressed by a liner function. The glass dissolution rate at the end time is determined by the LHS method as shown in Fig.1. The probability density functions of the glass dissolution rate and the end time are assumed to be a log-normal distribution and a uniform distribution, respectively.
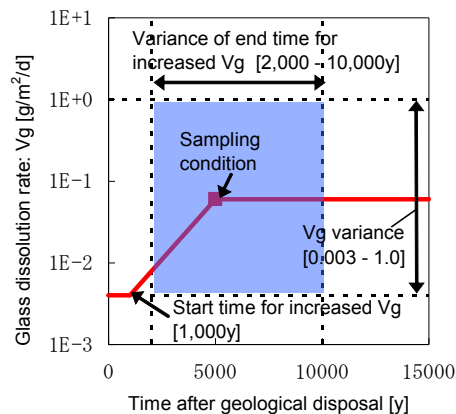

Fig.1 Time-dependence of glass dissolution rate for alteration scenario of the buffer material (The glass dissolution rate at the end time is determined by the LHS method.)

(3) Effective diffusion coefficient in the buffer material

We estimate the uncertainty on the effective diffusion coefficients of Se and Cs in the buffer material through the statistical analysis for previous experimental data of the effective diffusion coefficient. The appropriate data are picked up based on the range of the density specification of montmorillonite gel. Additionally, under the assumption that the effective diffusion coefficient of Se may depend on an ionic strength, the data of Se are selected from the standard level of ionic strength for a fresh type groundwater. Table2 shows the estimated variation of the effective diffusion coefficients of Se and Cs, which are corrected to the temperature. This variation is used for the analysis of the normal scenario.

In the alteration scenario, since the dissolution of montmorillonite may lead to decreased density of montmorillonite gel, the effective diffusion coefficients of Se and Cs is likely to increase. Because of few data for bentonite alteration, the minimum values for Se and Cs are respectively based on the average values in the statistical analysis mentioned above, and the maximum values are conservatively given by the effective diffusion coefficients in free water. The probability density function of effective diffusion coefficient is assumed to be a log-normal distribution. The time-dependence of effective diffusion

coefficient is basically treated in the same consideration mentioned in the selection of glass dissolution rate. In the analysis for the alteration scenario, it is assumed that the increase in effective diffusion coefficient starts at 1,000 y after the disposal (t=0) as the alteration progresses and the end time for increased diffusion coefficient is in the variation of 2,000 to 10,000 y. The relation between the effective diffusion coefficient and time from the start to the end time is assumed to be expressed by a liner function.

Table2 Variation of effective diffusion coefficient used in the calculations for normal scenario and alteration scenarios

parameter	scenario	element	unit	minimum[*1]	maximum[*1]
Diffusion coefficient in the buffer	Normal scenario	Cs	m^2/s	3E-11	1E-9
		Se		6E-12	1E-10
	Alteration scenario	Cs		2E-10	4E-9
		Se		2E-11	4E-9

(*1)The minimum and maximum values are treated as the values of 0.1 percentile and 99.9 percentile in a log-normal distribution.

(4) Distribution coefficient in the buffer material

The uncertainties on distribution coefficients in the buffer material are also estimated using the result of statistical analysis of them from previous experimental data. The variation of distribution coefficient is determined on the basis of the statistic, i.e. the average of measurements for compacted bentonite obtained from the in-diffusion method and the deviation of measurements from batch sorption tests. Moreover, the sorption data of Se are restricted to the reducing environment. Table3 indicates the variation of distribution coefficients of Cs and Se for the normal scenario.

The sorption property in the alteration of the buffer materials may deteriorate due to the dissolution of montmorillonite and mineralogical transformations. The sorption of the elements on the secondary minerals is conservatively ignored, and it is assumed that the variations of distribution coefficients for the alteration scenario are 0.1 times as low as those for the normal scenario. The probability density function of distribution coefficient is assumed to be a log-normal distribution. The time-dependence of distribution coefficient is treated in the same consideration mentioned above. In the analysis for the alteration scenario, it is assumed that the decrease in distribution coefficient starts at 1,000 y after the disposal (t=0) as the alteration progresses and the end time for decreased distribution coefficient is in the variation of 2,000 to 10,000 y. The relation between the distribution coefficient and time from the start to the end time is assumed to be expressed by a liner function.

Table3 Variation of distribution coefficient used in the calculations for normal scenario and alteration scenario

parameter	scenario	element	unit	minimum[*1]	maximum[*1]
Distribution coefficient in the buffer	Normal scenario	Cs	m^3/kg	1E-3	1E+0
		Se		1E-3	3E-2
	Alteration scenario	Cs		1E-4	6E-2[*2]
		Se		1E-4	3E-3

(*1)The minimum and maximum values are treated as the values of 0.1 percentile and 99.9 percentile in a log-normal distribution.
(*2)Logarithmic mean from the statistical analysis of Cs sorption data.

(5) Pore velocity in the buffer material

Under the assumption of the deterioration of hydraulic conductivity due to the long-term alteration, the pore velocity in the altered buffer for the case of changing into advective transport is estimated from the 2-D groundwater flow and trajectory analysis for the buffer material and the near rock mass. The illustration of the calculation is shown in Fig.2. The hydraulic conductivity of the hypothetical rock mass is considered to be in the range of 4E-11 to 2E-6 m/y on the basis of the variation of the measured data for the unaltered rock of high quality in Japan. We also assume the hydraulic conductivity of 0.01 and conservatively deterministic hydraulic conductivity of 1E-5 m/s of the altered buffer under montmorillonite dissolution and no mineralogical transformation. From the results of velocity analysis, the pore velocity in the altered buffer for the analytical case of advective transport is assumed to be in the variation of 1E-4 to 1.0 m/y. The probability density function of pore velocity is assumed to be a log-normal distribution. The time-dependence of pore velocity is treated in the same consideration mentioned above. It is assumed that the increase in pore velocity starts at 1,000 y after the disposal (t=0) as the alteration progresses and the end time for increased pore velocity is in the variation of 2,000 to 10,000 y. The relation between the pore velocity and time from the start to the end time is assumed to be expressed by a liner function.

The pore velocities in the analytical cases for normal scenario and for the alteration scenario of keeping diffusive transport are considered as the result of velocity analysis under the hydraulic conductivity of 1E-12 m/s of the buffer material.
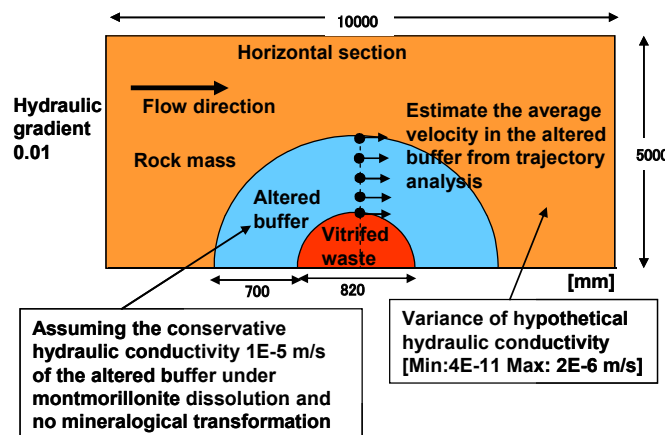

Fig.2 Illustration of the 2-D groundwater flow and trajectory analysis to estimate the variation of pore velocity in the case of advective transport in altered buffer

RESULTS AND DISSCUSSIONS

We carried out the uncertainty and sensitivity analysis for 1,000 parameter sets with LHS method. Fig.3 (a) shows the correlation between the maximum release rate of Cs-135 and the parameters in the case of diffusive transport in the altered buffer (CaseB1). The correlations between the Cs-135 release rate and two time-dependent parameters, effective diffusion coefficient and distribution coefficient of Cs, are not clear, but strong dependence on glass dissolution rate is indicated in Fig.3 (a).

Fig.3 (b) shows the comparison among the results of Cs-135 for the normal scenario (CaseA) and for three analytical cases (CaseB1, CaseB2 and CaseB3) of the alteration scenario. The 97.5[th] percentile of maximum release rate from the buffer material corresponding to the upper endpoint of the 95th percentile

confidence interval is applied to an index for comparing those calculation results.
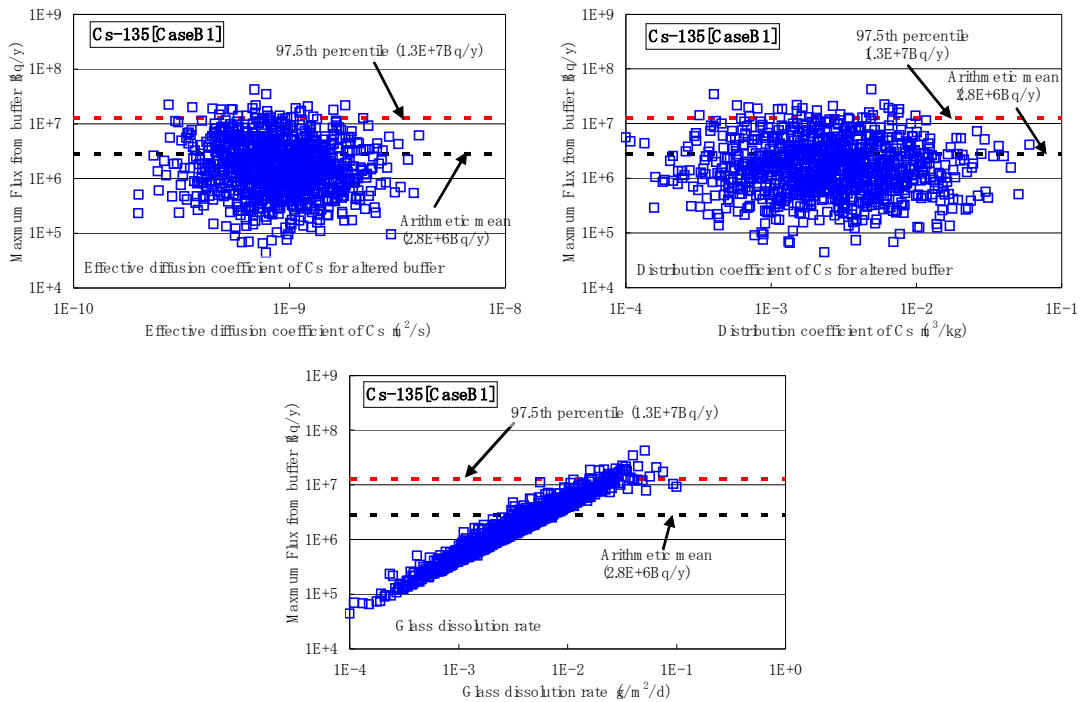
(a) Correlation between the maximum release rate and the parameters in CaseB1
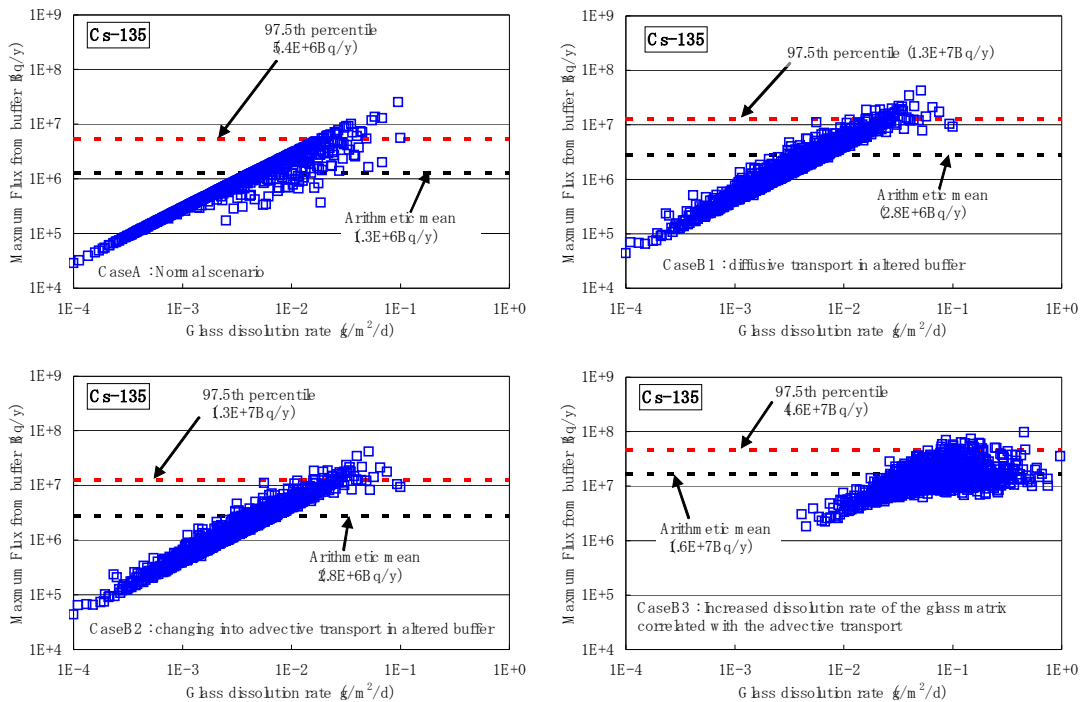
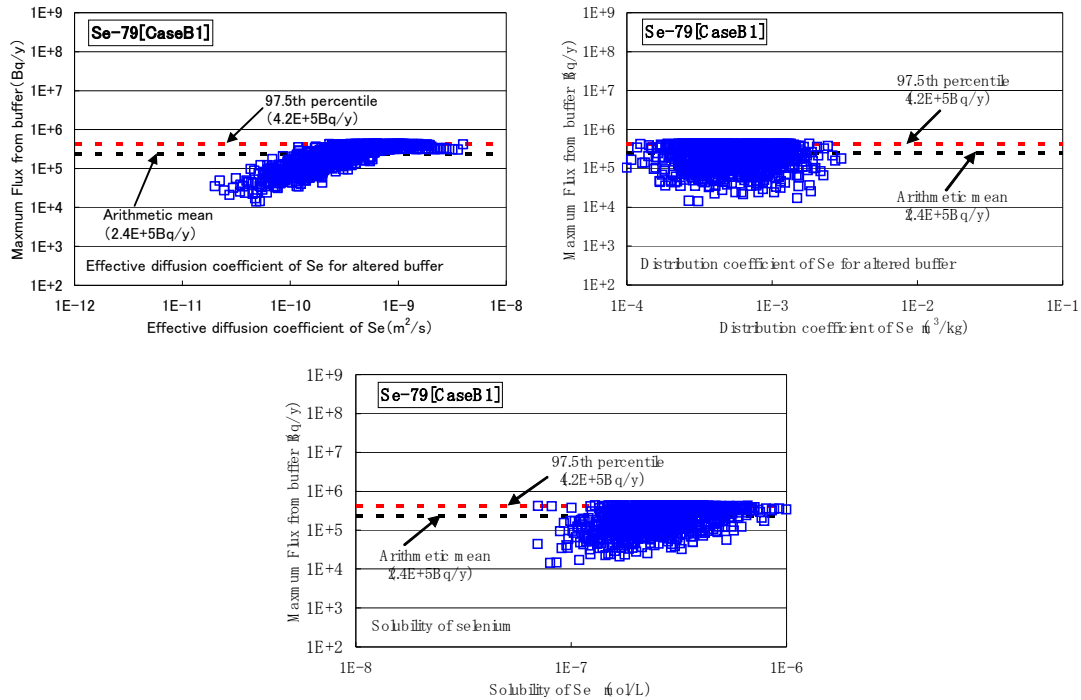(b) Comparison among the normal scenario and the alteration scenarios

Fig.3 Results of uncertainty and sensitivity analysis for Cs-135. (a) Correlation between the maximum release rate of Cs-135 and the parameters in the alteration scenario (CaseB1) and (b) Comparison among the results of Cs-135 for the normal scenario (CaseA) and the alteration scenarios (CaseB1,

B2 and B3)

The 97.5[th] percentile value in the case of diffusive transport in the altered buffer (CaseB1) is about twice as high as one in the case of normal scenario (CaseA). The time–dependence of two parameters, increased effective diffusion coefficient and decreased distribution coefficient of Cs with time, brings about a slight increased release rate of Cs-135 in CaseB1. There is no difference between the 97.5[th] percentile values in the cases of diffusive transport (CaseB1) and advective transport (CaseB2). Under the assumption that the long-term alteration of buffer have no correlation the increase in glass dissolution rate, these results indicate that the time–dependence of three parameters, i.e. pore velocity, effective diffusion coefficient and distribution coefficient of Cs, would only make a small effect on the maximum release rate of Cs-135 from the altered buffer. However, the 97.5[th] percentile value in CaseB3 is about one order of magnitude higher than in the case of normal scenario. Therefore, the assumption of increased the glass dissolution rate due to advective transport in the altered buffer would make a large effect on the maximum release rate of Cs-135 from the altered buffer.

The result of uncertainty and sensitivity analysis for Se-79 in the case of diffusive transport in the altered buffer (CaseB1) is shown in Fig.4 (a). The variation of three parameters, solubility of Se, increased effective diffusion coefficient and decreased distribution coefficient of Se in altered buffer, are considered in CaseB1. The results in CaseB1 indicate that the maximum release rate of Se-79 has a dependence on the solubility and the increased effective diffusion coefficient accompanied with the alteration of the buffer material. The correlation between the maximum release rate of Se-79 and the decreased distribution coefficient of Se is not fully recognized in Fig.4 (a). There is a limiting level of the maximum release rate of Se-79 in the scatter plot of Fig.4 (a). The reason is that the limit of release rate from the buffer material is determined by the dissolution rate and surface area related to dissolution of the vitrified waste form even if it happens to increase the release rate due to the variation of the other parameters.

Fig.4 (b) shows the comparison among the normal scenario and the alteration scenario for Se-79. The 95th percentile value in the case of diffusive transport in the altered buffer (CaseB1) is about six times as high as in the normal scenario (CaseA). The 95th percentile value in the case of advective transport in the altered buffer (CaseB2) is almost equivalent to in CaseB1. Moreover, the 95th percentile value in the case of increased glass dissolution rate correlated with the advective transport in altered buffer (CaseB3) is about twenty times as high as in CaseA. Three time-dependent parameters correlated with the alteration, i.e. pore velocity, effective diffusion coefficient of Se and glass dissolution rate make a large effect on the maximum release rate of Se-79. This result suggests the need to understand the feasibility on the scenario of increasing the glass dissolution rate accompanied with the alteration of buffer.

(a) Correlation between the maximum release rate and the parameters in CaseB1
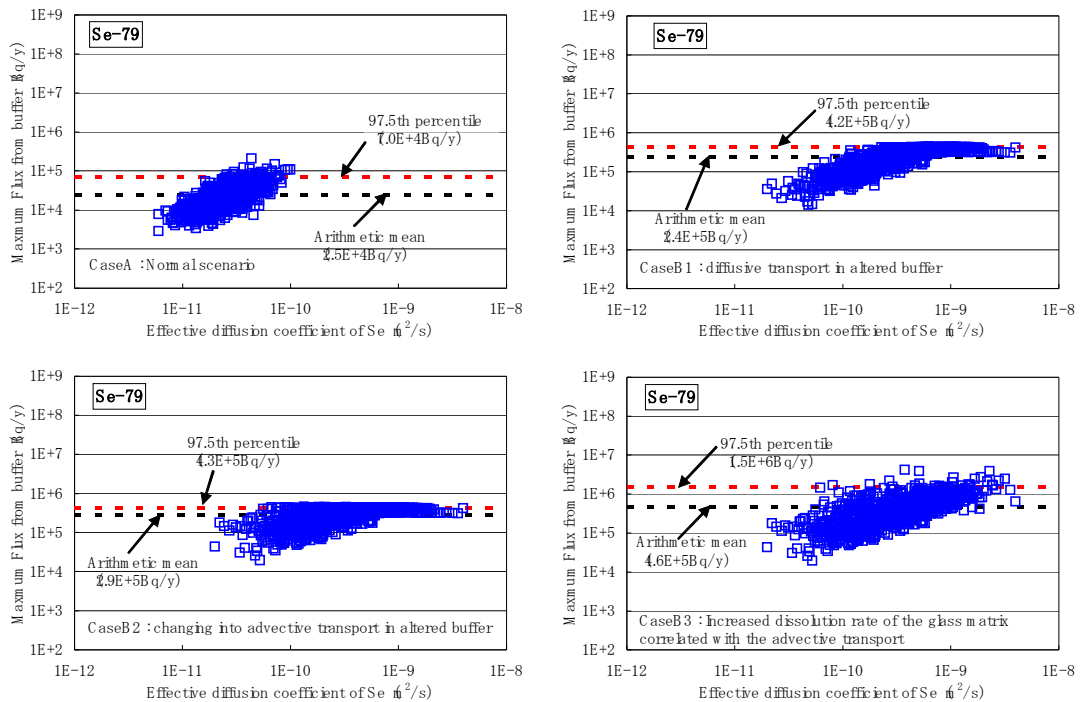
(b) Comparison among the normal scenario and the alteration scenarios

Fig.4 Results of uncertainty and sensitivity analysis for Se-79. (a) Correlation between the maximum release rate of Se-79 and the parameters in the alteration scenario (CaseB1) and (b) Comparison among the results of Se-79 for the normal scenario (CaseA) and the alteration scenarios (CaseB1, B2 and B3)

CONCLUSIONS

   Monte Carlo-based uncertainty and sensitivity analysis was carried out in order to identify important scenarios and parameters with respect to the long-term alteration of buffer material. In the possible scenarios of increased glass dissolution rate due to advective transport in the altered buffer, the maximum release rates of Cs-135 and Se-79 from the buffer material to the surrounding host rock increase conspicuously and are about ten (Cs-135) and twenty times (Se-79) as high as in the normal scenario, respectively. This increase in the maximum release rate of Cs-135 in the alteration scenario was caused by increased pore velocity and increased glass dissolution rate. Three time-dependent parameters correlated with the alteration, i.e. pore velocity, effective diffusion coefficient of Se and glass dissolution rate were sensitive to the maximum release rate of Se-79. Decreased distribution coefficients of Cs and Se caused by the alteration were insensitive parameters.

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