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Migration Behavior of Cesium Released from Fully Radioactive Waste Glass in Compacted Sodium Bentonite

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POWER REACTOR AND NUCLEAR FUEL DEVELOPMENT CORPORATION

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Migration Behavior of Cesium Released from Fully Radioactive Waste Glass in Compacted Sodium Bentonite

ASHIDA Takashi[1], KOHARA Yukitoshi[1], SHIBUTANI Tomoki[1], YUI Mikazu[1]

ABSTRACT

Migration experiment with compacted sodium-type bentonite saturated with distilled water was carried out at room temperature in order to investigate the coupled processes of Cs release from fully radioactive waste glass and its transport in compacted bentonite. Under these conditions, leaching of Cs from the waste glass and diffusion and sorption of Cs in compacted bentonite occur simultaneously.

Cesium migration behavior was analyzed by geochemical and transport modelling. The input data for this analysis were derived from the following individual measurements and models:

(a) The leaching behavior of Cs from the waste glass was analyzed by congruent dissolution with silica before its saturation and subsequently with boron.

(b) The diffusion coefficient of Cs in bentonite porewater was estimated from an electrochemical interaction model with the bentonite surface.

(c) The distribution coefficient of Cs was estimated from a cation exchange model.

The individual models were applied to compacted bentonite with consideration of the porewater chemistry, microstructure and surface characteristics of the bentonite and the solid/liquid ratio. The modelling results show a good agreement with the experimental observations.

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実高レベル放射性廃棄物ガラス固化体から放出された セシウムの圧縮ベントナイト中の移行挙動

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要　　旨

実高レベル放射性廃棄物ガラス固化体から放出されるCsの複合挙動および圧縮ベントナイト中の移行挙動を調査するため、蒸留水で飽和されたナトリウム型圧縮ベントナイトを用いた移行実験を室温において実施した。これらの試験条件下では、Csのガラス固化体からの浸出や圧縮ベントナイト中の拡散および収着過程は同時に起こるであろう。

Csの移行挙動は地球化学および移行モデルを用いて評価した。モデル計算に必要な入力データは、それぞれの挙動を独立させた個別の実験およびモデルから導いた。浸出挙動は、浸出液中のSi濃度が飽和に到達するまではSiとの調和溶解を、飽和後はBとの調和溶解を仮定することにより推定された初期浸出速度および残存溶解速度を与えた。ベントナイト間隙水中の拡散係数は、ベントナイト表面に対する電気化学反応モデルにより評価した。また、分配係数の評価には、イオン交換モデルを用いた。さらに、それぞれのモデルには、溶液化学、ベントナイトの微小構造や表面特性および固液比が考慮され、圧縮ベントナイトに適用された。モデルを用いた計算結果は実測値とほぼ一致するものであった。

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CONTENTS

1. INTRODUCTION

In geological disposal of high-level radioactive waste (HLW), radionuclides may be leached from the waste glass and migrate into the surrounding bentonite. Previous studies of the migration behavior of radionuclides in the bentonite have supplied data on the individual processes such as glass dissolution (e.g. Miyahara et al., 1990), diffusion (e.g. Sato et al., 1992) and sorption (e.g. Shibutani et al., 1994). Since these processes occur simultaneously in the bentonite, coupled behavior should be evaluated to confirm the reliability of individual models and data for the performance assessment of a high-level radioactive waste repository.

This paper presents the results of the experiments on Cs redistribution from fully radioactive waste glass in contact with compacted sodium bentonite. In fact, many radionuclides may be transfer to the surrounding bentonite from the waste glass as a result of leaching. The reason for which shown an interest in Cs among them because it is a great importance of the soluble elements. Though previous studies (Ashida et al., 1994) have reported on the migration behavior of Pu released from Pu-doped glass in compacted bentonite, the results were interpreted using empirical parameters. In this study, Cs migration was interpreted using more fundamental parameters based on a mechanistic understanding of its migration behavior. These parameters were obtained through the individual measurements. Leaching of Cs from the waste glass in the absence of the bentonite was measured by MCC-1 and MCC-3 tests. Diffusion behavior was estimated by consideration of an electrochemical interaction model between Cs ions and the bentonite surface. Sorption behavior and porewater chemistry were estimated mainly from a cation exchange model.

2. EXPERIMENTS

2.1 Preparation of Fully Radioactive Waste Glass

The high-level liquid waste (HLLW) for vitrification was generated from the Tokai Reprocessing Plant. The HLLW was mixed with a glass frit in a ceramic melter at 1150℃ for 2 hours. The molten glass was poured into a stainless canister with size of 60mm in diameter and 700mm in height. After gradual cooling to room temperature to prevent the formation of cracks, the homogeneity of the glass was verified by gamma scanning spectroscopy. The block was then cut and cored to produce specimens with size of ϕ20mm × H3mm, weighing approximately 2.5g and containing approximately 125 GBq of radioactivity per specimen. The specimens were washed using ethanol and dried at 90℃ for one hour. The composition of fully radioactive waste glass used for this experiment is shown in **Table I.** The details of the glass preparation are given elsewhere (Miyahara et al., 1989).

2.2 Leaching Experiments

Leaching experiments on fully radioactive waste glass were carried out using the MCC-1 and MCC-3 methods (MCC, 1981) with double-distilled water at room temperature (around 27℃) under aerobic conditions.

Static MCC-1 type experiments using glass cylinders were carried out from 14 days to 1140 days. The ratio of surface area to leachant volume (SA V^{-1}) was 10 m^{-1}. The weight of the glass specimen was measured before and after the experimental period. These experiments were carried out in a stainless steel reaction vessel coated with a gold layer 45μm thick; the specimens were supported in a platinum basket during leaching.

Leaching experiments similar to the MCC-3 method were carried out using powdered specimens in a closed, agitated system. The experimental period lasted from 28 days to 222 days. The glass powder was prepared by crushing. The particle size was under 840μm . The ratio of the weight of the glass specimen to the leachant volume was 100g l^{-1} : SA V^{-1} was at least 261 m^{-1} given that the powdered specimen was less than 840μm in diameter and the density of the waste glass was 2.74×10^3 kg m^{-3}. A stainless steel reaction vessel was also used in this experiment. The glass and the leachant were put in the reaction vessel and agitated 15 times per minute.

At the end of the experiment, the leachate was filtered through a 10,000 molecular weight cut-off (MWCO) ultrafilter. The pH was measured in the filtrates, the concenrations of Si and B were analyzed by direct coupled plasma spectroscopy (DCP) and the activity of Cs by gamma spectrometry.

2.3 Coupled Experiments

The procedure adopted was similar to that of the in-diffusion experiment reported by Miyahara et al.(1991). The sodium bentonite used is KunigelV1® with the mineralogy listed in **Table II** (Ito et al., 1994), from Kunimine Industries Co. Ltd.(Japan). The bentonite was dried at 110℃ for one week and then compacted using a hydraulic press and a punching tool into a stainless steel cell 20mm in diameter and 20mm in height to produce dry densities of 500 kg m^{-3} and 1,000 kg m^{-3}, respectively. The samples were then immersed in distilled water to allow saturation. The water saturation was monitored by the measurement of the weight of the cell. It was confirmed to reach in a month and the samples were remained to immerse in water for finally eight weeks until use this experiment. A waste glass sample was then placed between two pieces of the saturated bentonite. **Figure 1** gives a schematic view of an experimental cell.

The coupled experiments were carried out for 15, 35, 110 and 300 days at room temperature (around 27℃) under aerobic conditions. During the experiments, the weight of the cell was measured at appropriate intervals to check for leakage and its decrease was not confirmed to exceed 0.1g. After the experimental period, the glass specimen was separated from compacted bentonite and the bentonite cylinder was cut into 0.5-2mm thick slices. Each slice was placed in 1M HNO_3 solution for a week at room temperature to extract radionuclides migrated in compacted bentonite. The activity of Cs in the 1M HNO_3 solution was measured by gamma spectrometry after filtration through a 5,000 MWCO ultrafilter. Detailed tests confirmed that between 40% and 60% of Cs was extracted to depend on the Cs concentration in the bentonite under these experimental conditions.

3. RESULTS AND DISCUSSION

3.1 Leaching Experiments

The results of the MCC-1 experiments are shown in **Figure 2**, which indicates the leaching behavior of Si, ^{137}Cs and pH as a function of time. The results of the MCC-3 experiments are shown in **Figure 3** for B, Si, ^{137}Cs and pH. The leaching behavior of Cs released from the waste glass in the experimental pH range was expected to be described by congruent dissolution with a first order reaction rate law and a residual reaction rate (Grambow, 1985) as follows:

$$\frac{da_{Cs}}{dt} = f_{Cs} \cdot \frac{SA}{V} \cdot \left\{ k_{Si} \cdot \left(1 - \frac{a_{Si}}{a_{Si}^*} \right) \right\} + f_{Cs}' \cdot \frac{SA}{V} \cdot k_{rB}, \tag{1}$$

where a_{Cs} and a_{Si} are the activity of the Cs^+ ion and $Si(OH)_4(aq)$ in the leachate, respectively, a_{Si}^* is the activity of $Si(OH)_4(aq)$ at equilibrium with $SiO_2(am)$, k_{Si} is the initial leaching rate of Si, k_{rB} is the residual reaction rate of B, f_{Cs} and f_{Cs}' is the molar weight fraction of Cs to Si and B in the waste glass, respectively, SA is the surface area of the specimen, V is the leachate volume and t is time. In the dilute system in these experiments, the activities of species, a_{Cs}, a_{Si} and a_{Si}^* are more or less equal to concentrations C_{Cs}, C_{Si} and solubility C_{Si}^*, respectively.

Using equation (1), the leaching behavior of Si, B and ^{137}Cs were calculated under MCC-1 and MCC-3 experimental conditions to secure any evidence of the leaching behavior of Cs described above. The initial leaching rate for Si measured for 14 days in the MCC-1 experiments was estimated to be 3.35×10^{-2} g m^{-2} day^{-1}, which was the mean value of total glass weight loss and normalized weight loss of Si. The residual reaction rate is determined after the Si concentration was saturated in the leachate. **Figure 3** shows that the Si concentration is already saturated at 91 days. The residual reaction rate of the waste glass derived from normalized weight loss of B between 91 days and 222 days in the MCC-3 experiments was 2.67×10^{-3} g m^{-2} day^{-1}. The solubility of $SiO_2(am)$ under each experimental condition was calculated using the solution chemistry in **Table III** measured during the final period in each experiment as the equilibrium concentration.

The predicted release of Si, B and ^{137}Cs based on equation (1) is shown in **Figure 2** and **Figure 3**. These figures indicate that equation (1) can interpret the release behavior of Cs from the waste glass.

3.2 Coupled Experiments

The ^{137}Cs concentration profiles from the coupled experiments are shown in **Figure 4** and **Figure 5** for dry densities of 500 kg m^{-3} and 1,000 kg m^{-3}, respectively. The two concentration profiles for ^{137}Cs were obtained in the left and right sides of compacted bentonite. The apparent diffusion coefficients of ^{137}Cs determined from the slopes of the curves after an experimental period of 15 days by the best fit of an error function (Muurinen et al., 1985) were estimated to be 2.3×10^{-11} m^2 s^{-1} and 1.2×10^{-11} m^2 s^{-1} for dry densities of 500 kg m^{-3} and 1,000 kg m^{-3}, respectively. These values are similar to the apparent diffusion coefficients of ^{137}Cs obtained from previous in-diffusion experiments (Sato et al., 1993). The plateau profiles for the periods of 110 days and 300 days at both dry densities indicate a breakthrough of ^{137}Cs.

The input parameters for release and transport modelling were derived from independent analysis for individual processes.

It is considered that Si diffusion in compacted bentonite is increased in the release of Cs from the waste glass when the leaching behavior of Cs is presumed to congruent dissolution with the glass matrix. This effect was assumed not so significant based on the R value (the ratio of forward leaching rate to steady-state diffusive mass transfer rate) proposed by Zavoshy et al.(1985) under the experimental conditions. The pH dependence of glass dissolution behavior in compacted bentonite was also considered using an exponential term of proton activity in the leaching rate equation (Knauss et al., 1990). The value for the initial leaching rate was several times higher than that of the MCC-1 test, but the effect on the Cs concentration profile in compacted bentonite was not assumed to be significant. The leaching rate of Cs from the waste glass was taken from the leaching experiments as mentioned above.

The existing transport code in porous media, RELEASE (PNC, 1992), was modified to be consistent with the experimental geometry. A schematic illustration of the transport model is given in **Figure 6**. In this model, the volume of each model cell is the same.

The concentration of Cs with time in each model cell is described as follows:

$$\left(\varepsilon_B + \left(1 - \varepsilon_B\right) \rho_{th} Kd_B\right) V_n \frac{dC_n}{dt} = \varepsilon_B S_{n-1} J_{n-1} - \varepsilon_B S_n J_n \ , \tag{2}$$

where, $V_n = S_G \Delta x$, (3)

$$S_{n-1} = S_n = S_G , \tag{4}$$

$$J_{n-1} = -D\frac{C_n - C_{n-1}}{\Delta x} , \tag{5}$$

and

$$J_n = -D\frac{C_{n+1} - C_n}{\Delta x} , \tag{6}$$

where, n is the model cell number, ε_B is the porosity of compacted bentonite, ρ_{th} is the theoretical density of the bentonite (2.7×10^3 kg m^{-3}), Kd_B is the distribution coefficient of Cs in compacted bentonite, V_n is the volume of a model cell, C_n is the Cs concentration, S_n is the cross-sectional area of the bentonite (S_0 is the cross-sectional area of the bentonite-glass boundary), J_n is the flux from n-th model cell to $(n+1)$-th model cell (J_0 is the flux from the glass), Δx is the thickness of a model cell and D is the diffusion coefficient of Cs in porewater .

The Cs concentration at the outer boundary was assumed to increase due to the Cs accumulation. The inner boundary condition was the leaching rate of Cs described in equation (1).

The bentonite-water interaction and sorption of Cs was predicted using a cation exchange model for sodium smectite. Since Sasaki et al.(1995) reported that the results of the bentonite-water interaction experiments were a good agrement with the modelling resulte using an ion exchange reaction of smectite and Shibutani et al.(1994) shown the agreement between the measured distribution coefficient of Cs in compacted bentonite and the calculated value by a cation exchange model under aerobic conditions. For prediction of porewater chemistry, equilibrium with SiO_2(am) and calcite, pyrite oxidation and $Fe(OH)_3$ precipitation were assumed. Equilibrium with CO_2 gas was also assumed due to consideration of the saturation process of the bentonite for about 8 weeks under aerobic conditions. The input value of *pe*, the relative electron activity was given as instead of redox potential, was fixed 4.0 according to measured redox potential under aerobic conditions. These assumptions are also supported by a validation study of the bentonite-water interaction model (Sasaki et al., 1995). The effect of a small amount of impurities such as chloride was assumed not to be significant. The cation exchange constants were taken from the literature (Shibutani et al., 1998). The sorption site varies with the dry density of the bentonite because porewater chemistry also varies with the ratio of the bentonite mass to the volume of porewater. The porewater chemistry and distribution coefficients are shown in **Table IV** as calculated using their thermodynamic

data (PNC, 1992) and geochemical code PHREEQE (Parkhurst et al., 1980).

The diffusion coefficient of Cs in porewater was predicted by the following equation (Sato et al., 1992 ; 1993).

$$Dp = \frac{\delta}{\tau^2} \cdot Do, \tag{7}$$

where Dp is the diffusion coefficient in porewater, Do is the diffusion coefficient in free water, δ is the constrictivity and τ^2 is the tortuosity; these values were taken from the literature (Shibutani et al., 1998). The constrictivity, which defines the effect of ion exclusion, can be estimated using an electrical double-layer theory (Sato et al., 1995). The constrictivity (δ) is defined as

$$\delta = \frac{1}{d} \int_0^d \frac{n_x}{n_b} dx, \tag{8}$$

where n_x is the concentration of species n at distance x from the solid surface, n_b is the bulk concentration of Cs and d is the interlayer spacing of sodium smectite.

Table V summarizes the input data for the modified RELEASE code used to interpret the results of the coupled experiments. The modelling results are also shown in **Figure 4** and **Figure 5** with the measured ^{137}Cs profiles. The modelling results are considered to be in good agreement with the experimental results. The slope of the profiles depends on the diffusion and distribution coefficients, while the height of the profiles mainly depends on the leaching rate. The results indicate good estimates of fundamental constants for the leaching rate and the diffusion and distribution coefficients.

The residual reaction rate of fully radioactive waste glass in this study is dominated by chemical reactions. In recent studies, Inagaki et al.(1994) suggest that the diffusivity of soluble elements in the glass is the most effective parameter for determining the dissolution behavior on the long term. Our leaching experiment results are interpreted using Inagaki's model. Understanding the dissolution mechanism of soluble elements is critical for increasing the reliability of the glass dissolution rate on the long term.

4. CONCLUSIONS

Static MCC-1 and agitated MCC-3 type experiments using fully radioactive waste glass were carried out with double-distilled water at room temperature under aerobic conditions. The experimental results indicated that the dissolution behavior of Cs from the glass was congruent with silica before its saturation and with boron after saturation. The initial leaching rate and the residual reaction rate of the glass matrix were estimated to be 3.35×10^{-2} g m^{-2} day^{-1} and 2.67×10^{-3} g m^{-2} day^{-1}, respectively.

Coupled experiments with the waste glass sandwiched in compacted sodium-type bentonite were carried out at room temperature under aerobic conditions.

The Cs migration behavior was analyzed using a modified transport code. The input data for this analysis were derived from individual measurements and models. The agreement between the measured concentration profiles for ^{137}Cs and the modelling results indicates good estimates of fundamental constants for the leaching rate and the diffusion and distribution coefficients from the individual independent models.

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7. TABLE AND FIGURE LISTS

Figure 5 Measured concentration profiles for ^{137}Cs in compacted bentonite at experimental periods of 15 (○), 35 (□), 110 (▽) and 300 (◇) days and modelling results for a dry density of 1,000 kg m^{-3}. Calculated concentration profiles are represented as lines (a) - (d) for each period.

Figure 6 Schematic illustration of the modified RELEASE code. In the figure, inner (a) and outer (b) boundary conditions are shown; in (b), B and R represent bentonite and rock, respectively, and other symbols refer to equations (2) - (6) in text.

Table I Composition of fully radioactive waste glass

Element [1)]	Content (wt.%)	Radionuclide [1)]	Content (wt.%)
Si	2.46×10^{1}	^{239}Pu	4.27×10^{-2}
B	5.48×10^{0}	^{240}Pu	2.25×10^{-2}
Li	9.86×10^{-1}	^{241}Am	6.72×10^{-2}
Ca	1.28×10^{0}	^{244}Cm	2.35×10^{-3}
Zn	2.26×10^{0}	^{242}Cm	1.60×10^{-6}
Cr	1.08×10^{-1}	^{125}Sb	1.16×10^{-4}
Ni	1.15×10^{-1}	^{134}Cs	1.26×10^{-3}
Pd	2.86×10^{-1}	^{137}Cs	1.27×10^{-1}
Sm	1.62×10^{-1}	^{144}Ce	6.28×10^{-4}
Others [2)]	Not detected	^{144}Pr	2.65×10^{-8}
		^{154}Eu	4.21×10^{-3}
		^{155}Eu	2.04×10^{-4}
		Others	Not detected

1) Elements were analyzed by DCP and radionuclides were analyzed by alpha and gamma spectrometry.
2) Others include Na and Al.

Table II Mineralogy of KunigelV1®

Mineral	Content (wt.%)
Na-Smectite	46 – 49
Quartz	0.5 – 0.7
Chalcedony	37 – 38
Plagioclase	2.7 – 5.5
Calcite	2.1 – 2.6
Dolomite	2.0 – 2.8
Analcime	3.0 – 3.5
Pyrite	0.5 – 0.7

Table III Measured solution chemistry at final experimental period and calculated solubility of SiO_2 (am) using the PHREEQE code under the MCC-1 and MCC-3 experimental conditions

	MCC-1	MCC-3
Final experimental period	1140 days	222 days
pH	7.0	8.7
Concentration (mol l^{-1})		
B	1.87×10^{-4}	8.88×10^{-3}
Na	3.66×10^{-4}	1.17×10^{-3}
Li	1.34×10^{-4}	4.18×10^{-3}
Al	4.60×10^{-4}	No measurement
Solubility of SiO_2 (am) (mol l^{-1})	1.95×10^{-3}	2.03×10^{-3}

Table IV Bentonite porewater chemistry and the distribution coefficient of Cs

Dry density	500 kg m^{-3}	1,000 kg m^{-3}
pH	9.41	9.54
pe	4.00	4.00
Concentration (mol l^{-1})		
Na	3.11×10^{-1}	8.05×10^{-1}
K	3.46×10^{-3}	8.96×10^{-3}
Ca	2.43×10^{-2}	4.83×10^{-2}
Mg	4.48×10^{-3}	1.16×10^{-3}
Fe	6.11×10^{-8}	8.23×10^{-8}
S	2.00×10^{-3}	2.00×10^{-3}
Si	3.01×10^{-3}	3.35×10^{-3}
Z [1)]	3.45×10^{-1}	8.94×10^{-1}
Distribution coefficient of Cs (m^3 kg^{-1})	2.89×10^{-1}	2.09×10^{-1}

1) Z is defined as an ion exchangeable capacity on the bentonite surface.

Table V Input data for the modified RELEASE code

Dry density	500 kg m^{-3}	1,000 kg m^{-3}
Inner boundary condition	First order reaction rate law with residual reaction rate	
Initial leaching rate of glass matrix	3.35×10^{-2} g m^{-2} day^{-1}	
Residual reaction rate of glass matrix	2.67×10^{-3} g m^{-2} day^{-1}	
Weight fraction of Cs in glass	3.16×10^{-3} (-)	
Diffusion coefficient of Cs [1)]	2.81×10^{-4} m^2 day^{-1}	2.78×10^{-4} m^2 day^{-1}
Distribution coefficient of Cs [1)]	2.89×10^{-1} m^3 kg^{-1}	2.09×10^{-1} m^3 kg^{-1}
Porosity in compacted bentonite [2)]	8.15×10^{-1} (-)	6.30×10^{-1} (-)

1) These parameters depend on the porewater chemistry at each dry density.
2) Porosity in compacted bentonite (ε) was calculated as follows: $\varepsilon = 1 - \rho / \rho_{th}$, where ρ and ρ_{th} are the dry density and the theoretical density of compacted bentonite, respectively.

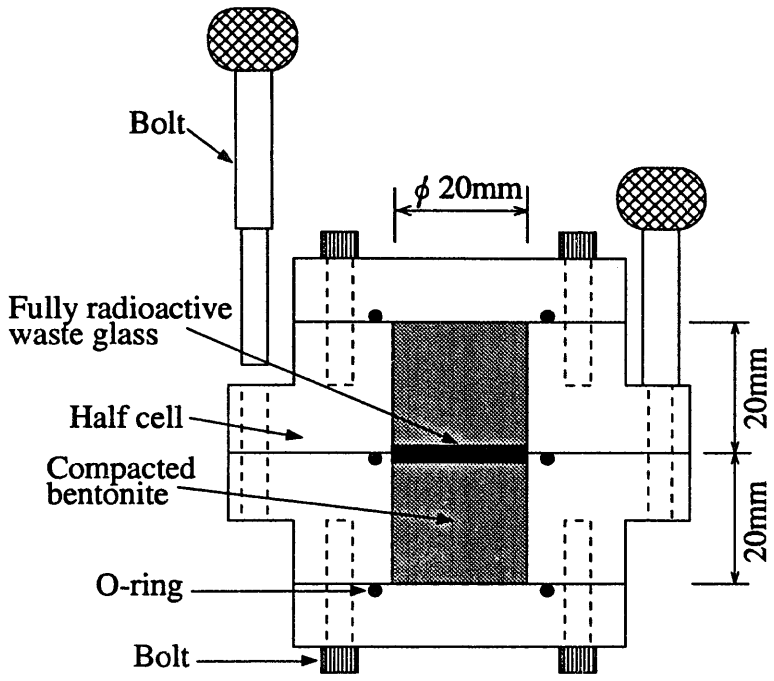

Figure 1 Schematic view of an experimental cell. During the contact of the waste glass with compacted bentonite, two half cells were joined via six bolts by remote manipulation: a good seal was achieved by using synthetic rubber for the O-ring.

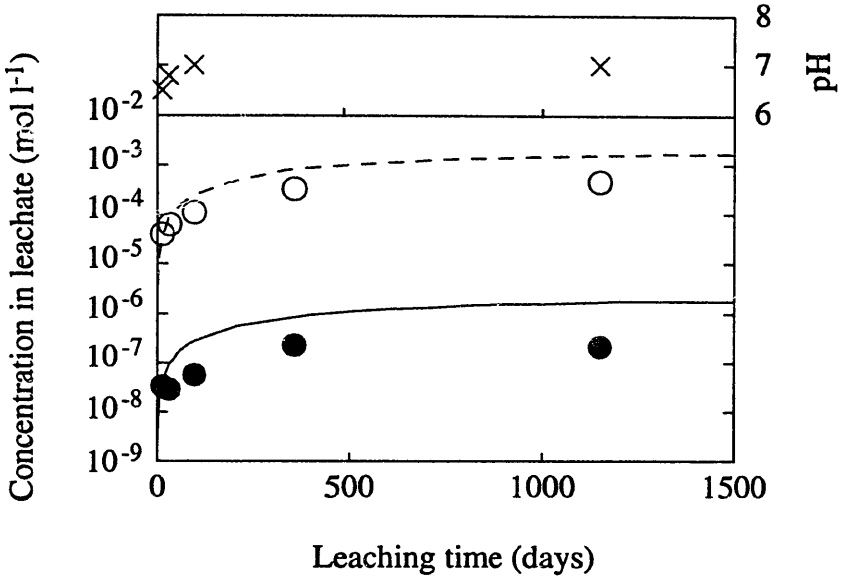

Figure 2 Results for ^{137}Cs (●), Si (○) concentration and pH (×) in leachate measured in the MCC-1 experiments and prediction of ^{137}Cs (solid line) and Si (dotted line) based on a first order reaction rate law.

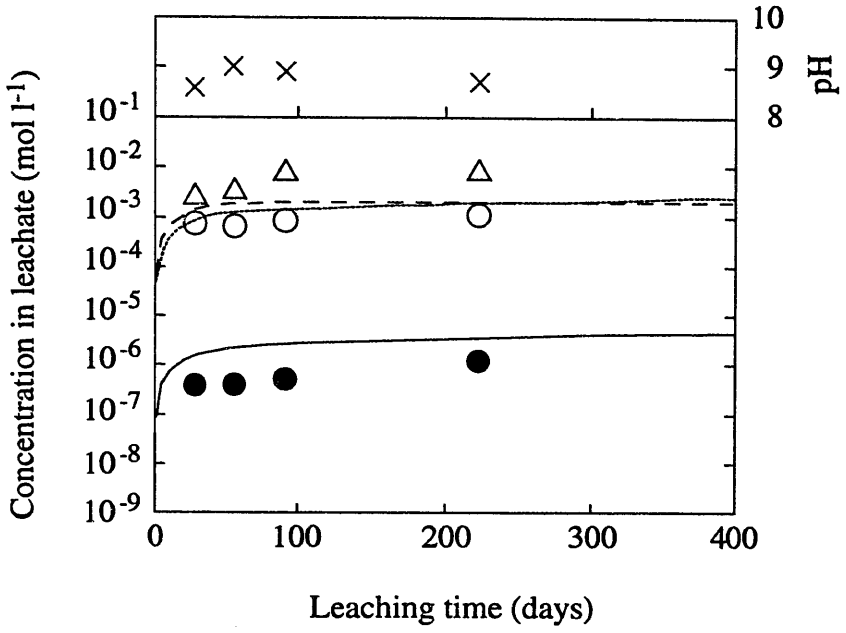

Figure 3 Results for ^{137}Cs (●), Si (○), B (△) concentration and pH (×) in leachate measured in the MCC-3 experiments and prediction of ^{137}Cs (solid line), Si (dotted line) and B (point line) based on a first order reaction rate law with a residual reaction rate.

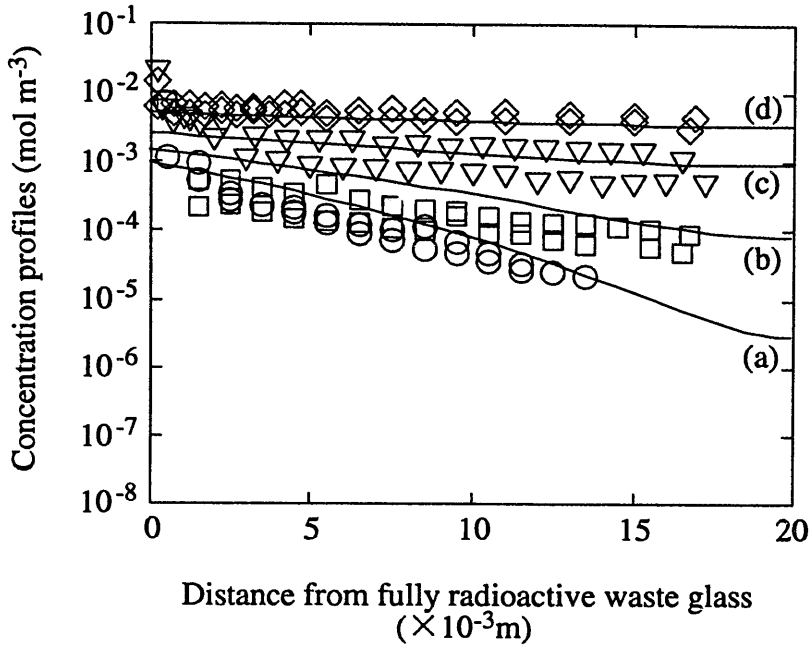

Figure 4 Measured concentration profiles for ^{137}Cs in compacted bentonite at experimental periods of 15 (○), 35 (□), 110 (▽) and 300 (◇) days and modelling results for a dry density of 500 kg m^{-3}. Calculated concentration profiles are represented as lines (a) - (d) for each period.

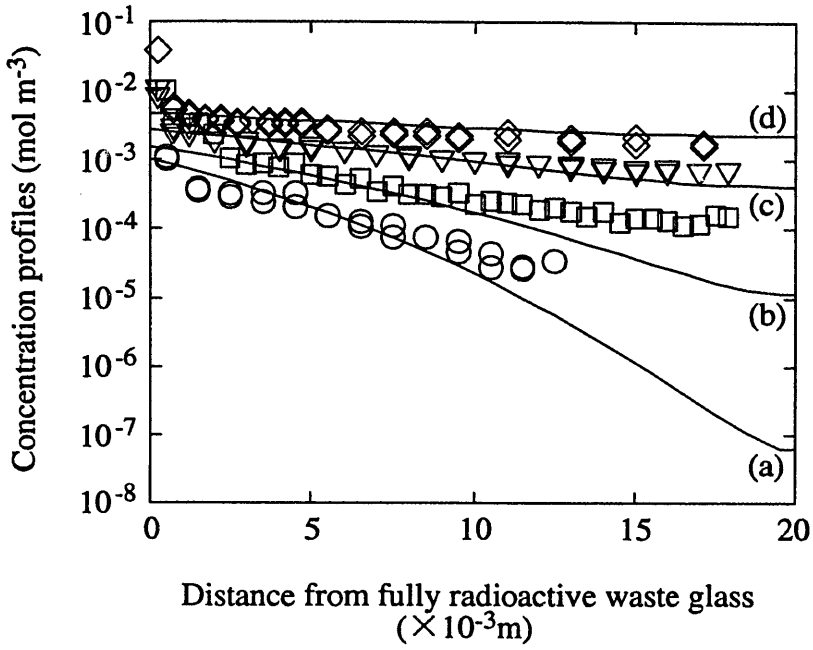

Figure 5 Measured concentration profiles for ^{137}Cs in compacted bentonite at experimental periods of 15 (○), 35 (□), 110 (▽) and 300 (◇) days and modelling results for a dry density of 1,000 kg m^{-3}. Calculated concentration profiles are represented as lines (a) - (d) for each period.

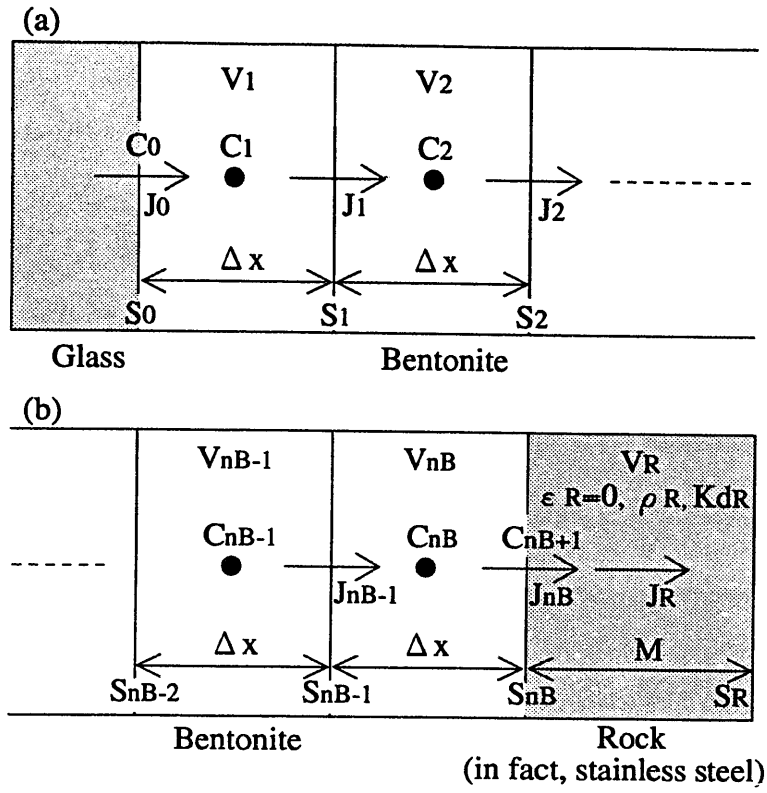

Figure 6 Schematic illustration of the modified RELEASE code. In the figure, inner (a) and outer (b) boundary conditions are shown; in (b), B and R represent bentonite and rock, respectively, and other symbols refer to equations (2) - (6) in text.