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Performance Assessment of Engineered Barrier for Retardation of Radionuclide Release in a Low– and Intermediate–Level Radioactive Waste Repository

W.J. Cho, J.O. Lee, P.S. Hahn, and H.H. Park
Korea Atomic Energy Research Institute
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중저준위방사성폐기물 처분장 인공방벽의 핵종유출 저지능 평가

조원진 · 이재완 · 한필수 · 박헌휘
한국원자력연구소
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Abstract

A simplified model to assess the performance of engineered barrier for the retardation of radionuclide release in a low– and intermediate–level radioactive waste repository was developed. The model is based on the repository design concept being suggested in Korea, and considers two types of release scenario ; a design–base release for the design of engineered barrier and a realistic release for the performance assessment. For the numerical illustration, the sample calculations were performed for five radionuclides with different chemical characteristics, and the results were analyzed.

요 약

중저준위 방사성폐기물처분장 인공방벽의 핵종유출 저지능을 평가할 수 있는 단순 모델이 제안되었다. 이 모델은 현재 우리나라에서 제안되고 있는 처분장 개념을 고려하여 고안되었으며, 인공방벽의 설계에 적합한 설계기준 유출 과 인공방벽 성능평가에 적합한 현실적 유출 두 가지 경우를 다루고 있다. 모델의 유용성을 보이기 위해 화학적 특성이 다른 다섯 핵종에 대해 모의 계산을 수행하고 그 결과를 분석하였다.

1. Introduction

In Korea a low– and intermediate–level radioactive waste repository is scheduled to be operated at the end of the 1990's, and the site investigation is under way. One of the principal issues arising from the implementation of radioactive waste disposal project is the disposal safety, which can be assured by the function of multi–barriers such as engineered barrier, geosphere, and biosphere. The performance assessment for an engineered barrier is considered to be important because it is closely related to the design of the repository, and it is allowed to control and predict its future behaviour.

The engineered barrier performance is classified into three groups ; mechanical stability, low hydraulic conductivity, and radionuclide retardation capability. The assessment for radionuclide retardation capability is especially important on the viewpoint of radiological safety for radioactive waste disposal, and depends on the design concept of the repository. In Korea, the radioactive wastes are appreciated to be disposed into rock caverns excavated in the host rock below ground surface. Although the detailed design concept of repository has not been defined yet, the preliminary design concept suggested that radioactive wastes should be disposed of, depending on their radioactivity, into one of three types of cavern ; Low-Level Waste (LLW) cavern for trash wastes, Solidified Concentrate Waste (SCW) cavern for solidified evaporator concentrates, and Intermediate-Level Waste (ILW) cavern for spent resins, spent filters and high activity trash wastes [1]. Both SCW cavern and ILW cavern may have backfill and similar design concept, while LLW cavern has however no backfill. As the SCW and ILW caverns contain the major part of radionuclide inventory in the repository, the assessment of radionuclide release rates from the SCW and ILW caverns is important on the viewpoint of radioactive waste disposal safety. According to the suggested design concept, the concrete wall will be installed to support stacked wastes in both SCW and ILW caverns, and the space between the concrete wall and the wall of cavern may be backfilled with clay or a crushed rock mixed with clay. Several studies have suggested the generic models to assess the radionuclide release rates from the high-level waste or the spent fuel repository [2,3]. In the present study, the methodology using simple mathematical models is suggested to assess the engineered barrier performance for retardation of radionuclide release from SCW cavern. Although developed for SCW cavern, the present methodology can be applied to the ILW cavern with minor modifications because both design concepts are similar each other except for the waste handling method that is not important in the assessment of the disposal safety.

2. System Description

The cross sectional view of SCW cavern considered in this study is shown in Fig.1 [1] The width, the height, and the length of carven are assumed to be 14.3 m, 11.7 m, and 200 m respectively, and the concrete boxes packed with waste drums will be emplaced inside the concrete wall with the thickness of 0.25 m. The interval between the concrete wall and the rock cavern wall is 0.9 m and the space is assumed to be backfilled with the mixture of the crushed rock and clay. After the closure of repository, the groundwater existing in the surrounding host rock intrudes into the cavern, and saturates the backfill. There are considerable empty spaces between each drum and between concrete wall and waste drums (void fraction is about 0.5) because of the cylindrical shape of waste drum, and the intruded groundwater will then fill in the voids. As time elapses, the waste container is corroded and the groundwater will eventually come into contact with cement-based waste matrix resulting in the radionuclide leaching into groundwater present in the void. The leached radionuclides will be transported into the surrounding backfill, and retarded for a considerable period as they return through the backfill, and finally released into surrounding host rock. The radionuclide release concept is represented schematically in Fig.2. Although the waste drums will be stacked uprightly in the real repository, they are represented as being stacked horizontally in the figure to enhance understanding the proposed release concept. The barrier effects of waste container and concrete wall are not considered here because they are less important relative to other engineered barriers. Dense clay-based materials have a low permeability [4],

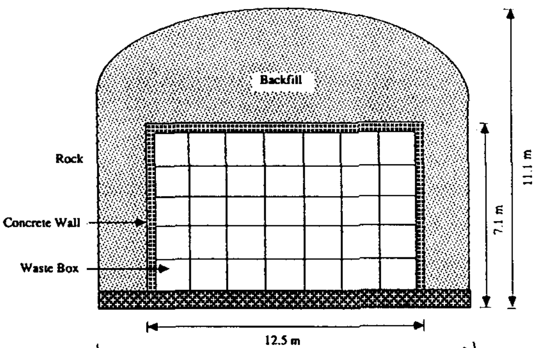

Fig. 1. Cross-Sectional View of the Suggested SCW Cavern [1]

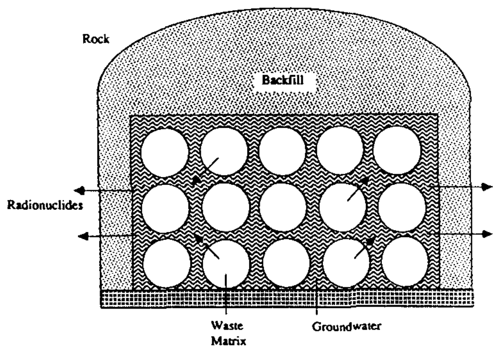

Fig. 2. Concept for the Radionuclide Release Through the Engineeredbarrier in the SCW Cavern

and moreover, the hydraulic gradient in a disposal cavern deep in hard rock is expected to be relatively low. Under these conditions, diffusion will be the principal mechanism of radionuclide release through backfill in the repository. For conservatism, it is assumed that the release occurs through the side wall of cavern at which the thickness of backfill is the smallest.

3. Mathematical Models

Considering the geometry of repository as shown in Fig.1, the radionuclide release through backfill can be approximated reasonably as the diffusional transport through a slab. It is also common practice to consider one-dimensional diffusion of radionuclide because the longitudinal diffusion is generally greater than transverse one.

Then the transient diffusional transport of radionuclide through backfill is given by the following equation.

$$\partial C_i/\partial t = D_{ai}\ \partial^2 C_i/\partial x^2 - \lambda_i\ C_i \qquad (1)$$

where

C_i : solution phase concentration of nuclide i

λ_i : decay constant of nuclide i

D_{ai} : apparent diffusion coefficient of nuclide i ($=D_{pi}/R_i$)

D_{pi} : pore diffusion coefficient of nuclide i

R_i : retardation factor of nuclide i

The retardation factor, R_i is defined as.

$$R_i = 1 + K_d \rho_d/\theta \qquad (2)$$

Here K_d is a distribution coefficient, θ is a porosity of backfill, and ρ_d is a bulk density of backfill.

The solution of Eq.(1) depends on the boundary and initial conditions. As there are no radionuclides immediately after the closure of repository, the initial condition is

$$C_i\ (x, 0) = 0 \qquad (3)$$

The outer boundary condition is replaced by an infinite medium boundary condition, and this approximation simplifies the mathematics considerably. The infinite medium is a good approximation because the transport rate of radionuclides past the interface between backfill and host rock is small [5,13,19]. The solution at the appropriate distance into the backfill is then taken to represent the concentration at the edge of the backfill, to serve as a source for migration through geosphere.

The inner boundary conditions depend on the degree of conservatism, and can be classified into two cases as follows.

Design-base Release

This is more conservative case which is suitable for the purpose of design of backfill in the repository. In this case, it is assumed that all radionuclides leached out from the waste matrix are transported into the backfill from the spot where they are leached out into the void water. The release rates of radionuclides therefore determine the amounts of nuclides transported into backfill, and the chemical equilibrium or solubility limit of radionuclides in the near-field groundwater are not considered. This approach over-estimates the radionuclide release rates from the backfill and thus can be applied to the design base scenario that has to consider the severe condition.

By continuity, the diffusive flux of nuclide at the inner boundary of backfill must be equal to the leach rate of nuclide from waste matrix. Then the boundary conditions are

$$[-\partial C_i/\partial x]_{x=0} = 1/D_{pi}\theta\,(A_w/A_b)\,[M_i(t)\exp(-\lambda_i t)] \quad \text{and} \qquad (4)$$

$$C_i(\infty, t) = 0$$

where $M_i(t)\exp(-\lambda_i t)$ is the leach rate per unit area. A_w and A_b are the total surface area of waste matrices and the inner side of backfill, respectively. Applying the Laplace transform technique to Eq.(1) with the initial and boundary conditions of Eq.(3) and (4), the following solution is obtained [7].

$$C_i(x,t) = [H_i \exp(-\lambda_i t)(D_{ai}/\pi)^{1/2}] \int \{M_i(t-t')/t'^{1/2} \times \exp[-x^2/(4t'D_{ai})]\}\,dt' \qquad (5)$$

where,

$H_i = 1/D_{pi}\theta(A_w/A_b)$

Although it is desirable to determine the leach rates from the experimental results obtained under the repository conditions with low water-to-waste matrix ratio, it is difficult to conduct the experiment under such conditions. The leach retes are therefore estimated from the short-term batch experimental results, and the use of batch experimental results gives over-estimates, but conservative leach rates. The leach rate models are based on the several types of waste geometry; infinite slab, semi-infinite slab, and finite cylinder etc. [8,9]. The semi-infinite slab assumption gives generally higher leach rate than finite cylinder geometry, but the difference is not significant because the nuclide diffusion coefficients through waste matrix are low [10]. Assuming semi-infinite slab, the radionuclide release rate is given by the following equation: [8,9]

$$M_i(t)\exp(-\lambda_i t) = J_i(t) = 2C_{owi}\,D_{awi}\exp(-\lambda_i t)/d \sum_{n=0}^{\infty}\exp[-(2n+1)^2 \times \pi^2 D_{awi}t/4d^2] \qquad (6)$$

where

D_{awi} : apparent diffusion coefficient of nuclide i through waste matrix

C_{owi} : initial nuclide concentration in waste matrix

d : half-thickness of waste matrix

For a special case that (a) there are no voids between the waste matrices and (b) total waste matrices are assumed to be equivalent to a single waste matrix with same volume, and (c) there is also no gap between waste matrices and backfill forming a double layer monolith, an analytical solution was given by Fraser and Javis [11]. It is however likely to under-estimate the radionuclide release rates from backfill and thus was not considered here.

Realistic Release

This is more realistic case applicable to the performance assessment of engineered barrier in the repository. It assumes that the leached radionuclide is transported into backfill at a rate that is controlled by the equilibrium concentration determined from chemical condition in the repository. For most nuclides, the leach rates from waste matrix to void water may be larger than diffusive fluxes from void water to backfill. Furthermore the presence of large amounts of cement/concrete in the repository increases the pH (up to 12–13), and decreases Eh resulting in strong base and reduction environment, and then decreases the equiliblium concentrations of radionuclides in groundwater [12].

The radionuclide concentration in repository is assumed to be maintained at a constant value determined by the chemical condition of groundwater/waste matrix/container system. It is also assumed that the consumed radionuclides by transport into backfill and radioactive decay are supplied by the immediate desorption of sorbed nuclides on cement, and the aqueous phase concentration is contant until all radionuclide inventory is consumed, and then drops to zero. For the band release, the boundary conditions are :

$$\begin{aligned} C_i(0,\ t) &= C_{oi} \qquad 0<t<T \\ &= 0 \qquad t>T \qquad (7) \\ C_i(\infty,\ t) &= 0 \end{aligned}$$

Applying the initial condition Eq.(3) and the boundary conditions Eq.(7) into Eq.(1), the solution is :

$$\begin{aligned} C_i/C_o = 1/2\, \{ & \exp[-x(\lambda_i/D_{ai})^{1/2}] \\ & \mathrm{erfc}[x/(2(D_{ai}t)^{1/2}) - (\lambda_i t)^{1/2}] \\ & + \exp[x(\lambda_i/D_{ai})^{1/2} \\ & \mathrm{erfc}[x/(2(D_{ai}t)^{1/2}) \\ & + (\lambda_i t)^{1/2}]\} \\ & [h(t)-h(t-T)] \qquad (8) \end{aligned}$$

where h(t) is a Heaviside function [13,14].

The change of radionuclide inventory is given by the following equation :

$$-dI_i(t)/dt = D_p\theta A(\partial c/\partial x)_{x=0} + \lambda_i I_i(t) \qquad 0<t<T_i \qquad (9)$$

where the diffusive flux of radionuclide at the interface of backfill and void water is given as : [14]

$$D_p(\partial c/\partial x)_{x=0} = -D_p/\sqrt{\pi D_a}\ C_o\ t^{-1/2} \qquad (10)$$

As $I_i(T_i)=0$, the band release time T_i can be obtained from Eq.(9).

4. Swedish Concept

The design concept of the intermediate–level waste cavern (BMA) in Swedish final repository for low– and intermediate–level waste (SFR) is similar to the suggested one for SCW cavern described in the previous section except for the backfill. In BMA, the space between the concrete wall and the host rock wall will be filled up with gravel, mainly to mechanically support the concrete walls [15]. The radioactivity is assumed to be evenly distributed inside the concrete construction, and the inside of construction is modelled as a stirred tank. The sorption equilibrium of radionuclide in the waste matrix is considered. The radionuclides are transported from the concrete construction into the void space between concrete wall and the wall of cavern by diffusion through concrete wall and then from the void space to the host rock by water flowing through gravel backfill in the cavern. The radionuclides and the water are delayed in the void space before release into host rock.

Therefore, the radionuclide release rates for Swedish concept were estimated from Eq.(8) and Eq.(9) with the values of D_{ai}, x, and θ for concrete.

5. Numerical Results and Discussion

For the numerical illustration of the present methodology, it was applied to calculate the radionuclide release rates from the backfill in the repository with the design concept described in the previous section. The disposal capacity of the repository was assumed to be 250,000 drums (as 200 L carbon steel drum). These are composed of 110,000 drums LLW, 70,000 drums SCW, and 70,000 drums ILW [1]. The release rates were calculated for five radionuclides; H-3, C-14, Ni-63, Sr-90, and Cs-137. These are principal nuclides for the safety assessment of low- and intermediate-level waste disposal because of their relatively large inventories and long half-lives. H-3 and C-14 are non-sorbing radionuclides, and Sr-90 and Cs-137 are strongly sorbing nuclides in the clay-based backfill. Ni-63 is moderately sorbed on clay. The radionuclide inventories shown in the Table 1 were estimated using the Sweden SFR data [16]. It was assumed that the 90 % of total inventories are contained in SCW and ILW. Furthermore the nuclide concentrations in SCW are taken to be the same as those in ILW. This assumption gives overestimation of the specific activity in SCW resulting in the conservative results. The concentration C_o in Eq.(7) for the realistic release should be obtained by considering the chemistry of the near-field system that is composed of waste matrix, concrete, carbon steel container, clay-based backfill material and groundwater. However, in this study, because of lack of informations, the C_o was calculated from the simple sorption equilibrium equation: $C_o = I_o / V[\theta + (1-\theta)\rho_s K_{dc}]$, where V is the waste volume, θ is porosity of waste matrix, ρ_s is the real density of waste matrix, K_{dc} is the distribution coefficient of nuclide on cement. Here, θ and ρ_s are taken to be 0.5 and 2700 kg/m^3, respectively. The sorption of radionuclide on cement depends, in a complex manner, on the ingredients of the cement-based waste matrix, pore-water chemistry, nuclide speciation etc.. In addition, sorption properties change with time due to chemical alternation. Here the K_{dc} (L/kg) values were conservatively assumed to be 0 for H-3, 1 for Cs-137 and Sr-90, and 100 for C-14 and Ni-63 [17,18]. To assess the backfill performance for the retardation of radionuclide release, the radionuclide release rates from the outer boundary of backfill were calculated as a function of time after the closure of repository. The values of input parameters used in the calculation are shown in Table 1 [8,9,13,16].

The radionuclide concentrations at the edge of backfill as a function of time are shown in Fig.3 to Fig.7, and those at the edge of concrete wall for Swedish concept are also represented. For Cs-137, the results for both the design base release and the realistic release are not shown because the concentrations are below 10^{-20} Ci/m^3 at all time, and thus can be negligible.

The results show that the radionuclide concentrations for design-base release are higher than those for realistic release at all time, and the degree of difference is influenced by sorption characteristics of radionuclides on clay-based backfill

Table 1. Input Parameters Used to Calculate the Radionuclide Release Rate From the Backfill

Radio-nuclide	λ_i (y^{-1})	I_0^{++} (C_i)	C_{ow} (C_i/m^3)	C_o (C_i/m^3)	D_a (m^2/y)	D_p (m^2/y)	D_{aw} (m^2/y)
H-3	5.63E-2$^+$	6.0E+3	1.9E-1	8.4E-2	6.3E-3	6.3E-3	1.0E-2
C-14	1.12E-4	1.6E+1	5.1E-4	1.9E-6	6.3E-3	6.3E-3	1.0E-5
Ni-63	6.93E-3	1.2E+4	3.9E-1	1.4E-3	4.7E-5	6.3E-3	1.0E-5
Sr-90	2.39E-2	2.4E+2	7.7E-3	1.6E-3	4.7E-5	6.3E-3	3.2E-4
Cs-137	2.29E-2	4.3E+3	1.4E-1	2.8E-2	9.5E-6	6.3E-3	1.6E-3

\+ 1.0E-1 means 1.0×10^{-1}

\+ + based on the disposal capacity of 250,000 drums

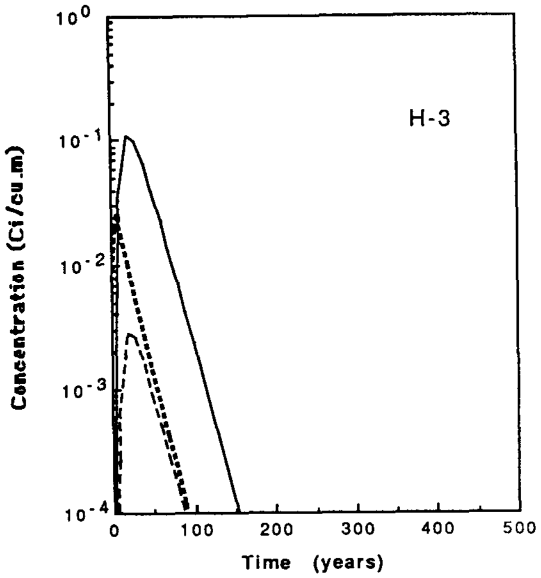

Fig. 3. Radionuclide Concentration in Groundwater at the Edge of Backfill for H-3 (——Design-Base Release······Realistic Release·······Swedish Concept)

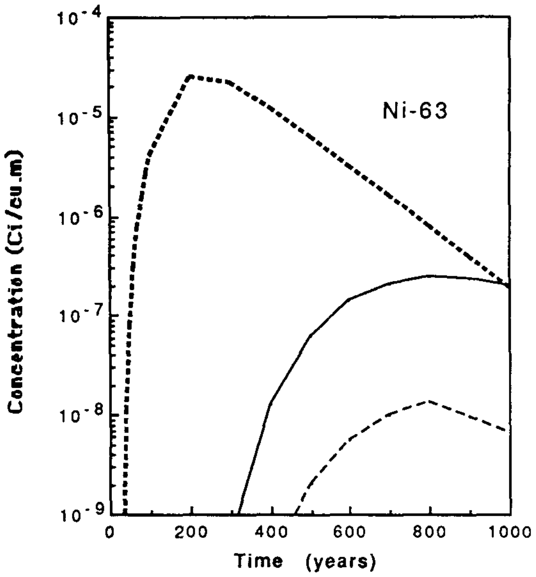

Fig. 5. Radionuclide Concentration in Groundwater at the Edge of Backfill for Ni-63 (——Design--Base Release······Realistic Release······Swedish Concept)

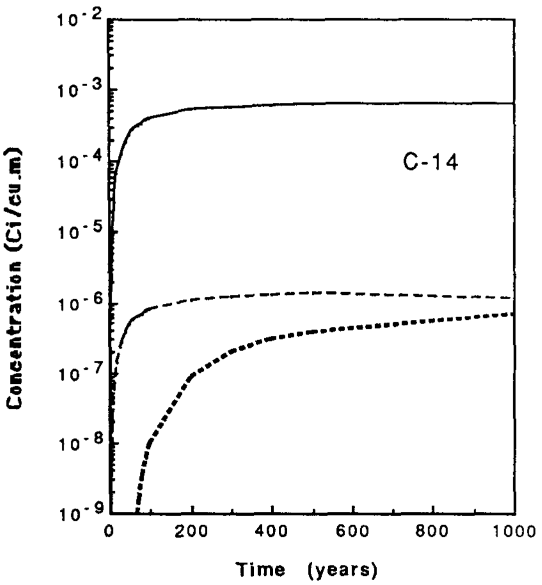

Fig. 4. Radionuclide Concentration in Groundwater at the Edge of Backfill for C-14 (——Design--Base Release······Realistic Release······Swedish Concept)

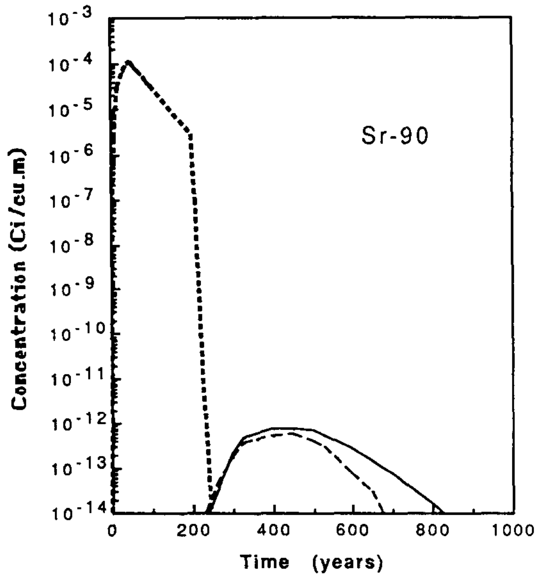

Fig. 6. Radionuclide Concentration in Groundwater at the Edge of Backfill for Sr-90 (——Design--Base Release······Realistic Release······Swedish Concept)

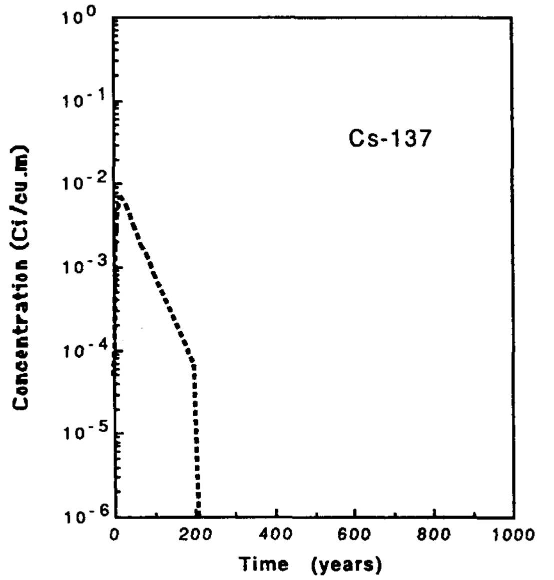

Fig. 7. Radionuclide Concentration in Groundwater at the Edge of Backfillfor Cs–137 (······Swedish Concept)

and cement. H–3 is a non–sorbing nuclide in both clay–based backfill and cement, and then the concentration depends on the influx of nuclides into backfill resulting in the higher values for design –base release. As C–14 is not sorbed on backfill, but sorbed strongly on cement, the release from backfill is entirely controlled by the influx rate into backfill from void water and the difference between the two releases is the largest. On the other hand, Sr–90 is sorbed strongly on clay–based backfill, but nearly not sorbed on cement, and the release controlling step is the diffusion through backfill so that the release rates are almost the same for both cases. Cs–137 showed similar behaviour to Sr–90. Ni–63 is sorbed on backfill moderately and on cement strongly so that it gives intermediate trend.

The results for the Swedish concept show somewhat different trends. For Cs–137, and Sr–90, the radionuclides are released at the early years and the concentrations are much higher than those from both the design base release and the realistic release. This is due to the fact that both radionuclides are strongly sorbed on clay but not sorbed on cement, thus the concrete wall is nearly not a barrier for Cs–137. Ni–63 is sorbed on both clay and cement, and higher concentrations for the Swedish concept are due to the small thickness of concrete wall as compared with clay–based backfill. As H–3 is a non–sorbing nuclide in both clay and cement and has a short half–life, the release rate for the Swedish concept is intermediate level. On the other hand, C–14 is strongly sorbed on cement, and the concrete wall is an effective barrier resulting in low concentration at all time despite of small wall thickness.

These results represent the mutual aid of concrete and clay to improve the performance of engineered barrier. Namely, the C–14 which is a non–sorbing nuclide in clay–based backfill is releaesd without retardation in the case of no cement, and will become one of the dominant nuclides in the long–term safety assessment. If the presence of cement is however considered, the risk from C–14 can be greatly reduced. As Sr–90 and Cs–137 are nearly not sorbed on cement but strongly sorbed on clay–based backfill, the barrier effect of backfill becomes important resulting in the negligible release. Therefore, the combination of cement and clay–based backfill can improve the effects of engineered barrier in the repository. However, the presented model is only preliminary one and further works on the following aspects are recommended to enhance its applicability; (1) determination of the radionuclide concentrations (C_o) in the near–field for the realistic release that is a complicate function of near–field chemistry, (2) analysis of the relative effects of the concrete wall, clay–based backfill, and the combinational use of concrete wall and clay backfill as a barrier to retard the radionuclide release.

6. Summary and Conclusions

The performance of engineered barrier in the repository was assessed using the simplified model. Two types of radionuclide release scenario, namely, more conservative one, and more realistic one were considered. The difference of radionuclide release rates in both cases is the largest for C–14, and the smallest for Sr–90 and Cs–137, depending on their chemical characteristics. The comparison between the results for the suggested design concept and Swedish design concept represents the mutual aid of concrete wall and clay–based backfill to retard the release of radionuclides, especially C–14, and Cs–137.

Therefore, the combinational use of clay–based backfill and concrete wall can enhance the performance of engineered barrier in the repository.

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