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A Conservative Safety Study on Low-Level Radioactive Waste Repository Using Radionuclide Release Source Term Model

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선원항 모델을 사용한 저준위 방사성폐기물 처분장의 보수적인 안전성고찰

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Abstract

A simplified safety assessment is carried out on rock–cavern type disposal of LLW using the analytical repository source term (REPS) model. For reliable prediction of the leach rates for various radionuclides, degradation of concrete structures, corrosion rate of waste container, degree of corrosion on the container surface, and the characteristics of radionuclides are considered in the REPS model. The results of preliminary assessment show that Cs–137, Ni–63, and Sr–90 are dominant. For the parametric uncertainty and sensitivity analysis, Latin hypercube sampling technique and rank correlation technique are applied. The results of the potential public health impacts show that radiological dose to intruder in the worst case scenario will be negligible and that more attention should be given to near–field performance.

요 약

암반동굴 타입의 저준위방사성폐기물 처분장의 보수적인 안전성평가를 처분장 선원항 REPS 모델을 사용하여 수행하였다. 신뢰할만한 핵종별 침출율 예측을 위하여 REPS 모델에서 콘크리트 구조물의 열하시간, 부식의 형태와 부식율, 드럼표면의 부식면적비, 그리고 핵종의 특성등이 고려되고 있다. 예비평가의 결과로 Cs-137, Ni-63, Sr-90등이 주요한 핵종임을 알 수 있다. 파라메타의 불확실성과 민감도분석을 위하여 라틴하이퍼큐브 샘플링과 Rank Correlation 기법이 사용되었다. 침입자 시나리오를 적용하였을 경우의 예상 피폭선량도 허용치 이하임과 처분장의 환경영향평가에 있어서 비교적 불확실성이 적은 Near Field의 중요성에 대한 인식이 새롭게 강조되어야 할 필요가 있음을 알 수 있었다.

1. Introduction

The cumulative amount of operational waste from nuclear power plants in Korea amounted to about 34,000 drums by the end of 1991 and will reach up to about 105,000 drums by 2000. A centralized repository for low-level radioactive waste (LLW) is scheduled to be constructed by the end of 1995 with an initial capacity of 250,000 drums. The subsequent expansions are also scheduled to increase the capacity eventually up to 1 million drums.

The potential public health impacts associated with rock-cavern type disposal of LLW have been preliminarily assessed to ensure that radiological doses to persons who might be exposed do not exceed any regulatory limits. The performance of far field is difficult to be predicted conclusively because of the high demands on site characterization. Considering the uncertainty regarding performance assessment of far field, it is desirable at this moment to analyze near-field performance for a preliminary performance assessment. In this paper, the results of sensitivity and uncertainty analysis of radionuclide release from engineered barriers in low-level radioactive waste repository are presented using Latin hypercube sampling and rank correlation techniques and the analytical repository source term (REPS) model[1].

2. Methodology

2.1. Process Description

The objective of the low-level radioactive waste repository source term model is to develop a system model capable of predicting radionuclide release rates from cemented waste drums disposed of in an underground repository. The repository source-term model is divided in three compartments : degradation of concrete structures, corrosion of waste container, and waste form leaching. Each of these compartments is described by submodels which will be coupled into the system model.

No penetration of groundwater may occur while the concrete structure retains its integrity. The degradation of a reinforced concrete depends on environmental characteristics of a specific disposal site. In this preliminary assessment, it is conservatively assumed that the degradation is represented by a parameter named degradation time. The degradation time is assumed to have a lognormal distribution as shown in Table 1.

2.2. Corrosion Model

The primary cause for a loss of containment ability of steel container is corrosion. Corrosion of the radioactive waste container may be predicted from mechanistic models or empirical models[2]. The corrosion model of REPS is empirical in nature and uses a corrosion data base. The general form of corrosion depth equation is

$$d = kt^n \tag{1}$$

where d is the corrosion depth, k is a corrosion rate constant from the corrosion data base, t is time, and n is an empirical constant which gives the time dependence of corrosion depth. In this paper, n is assumed to be 1, which is a conservative assumption for long-term corrosion depth prediction[3,4].

The degree of corrosion, C_R, that is defined as the ratio of the surface area of the waste form exposed to water to the total surface area of a container, is assumed to be expressed by the following experimental formula[5]

$$C_R(t) = \exp(\alpha + \beta t)/(1 + \exp(\alpha + \beta t)) \tag{2}$$

where α and β are empirical constants determined by fitting experimental data. Based on the extrapolation of a long-term corrosion experiment of 1.6mm thick drum[5], $\alpha = -5.3$ and $\beta = 0.12$ are assumed to simulate the gradual increase of corroded surface area on waste drum.

2.3. Leach Model

Leach rates, i.e., the rates at which radionuclides are released from the solid waste form into the contacting groundwater, constitute the source term to radionuclide hydrogeological transport models. The analysis is based on time-dependent leaching at constant temperature, appropriate for non-heat-generating low- and intermediate-level radioactive wastes. Three leaching mechanisms included in the REPS model are solubility-limited release, inventory-limited release,and solid diffusion controlled release[1].

The fractional release rate $f_i(t)$ as a function of time t after emplacement from a waste drum to surrounding porous medium is

$$f_i(t) = \frac{m_i(t\,;\,t_0,T_i)\,C_R(t)}{M_i^0} \tag{3}$$

where

$m_i(t\,;\,t_0,T_i)$ is the leach rate of radionuclide i from a waste form at time t after emplacement,

M_i^0 is the initial inventory of species i,

t_0 *is the container penetration time or the starting* time of leaching after emplacement,

and

T_i is the leach time or the duration of leaching for radionuclide i.

For three leaching models included in the REPS code, the leach rate $m_i(t\,;\,t_0,T_i)$ is as follows :

● Solubility-limited model

The solubilit-limited diffusive release rate of radionuclide i is

$$m_i(t\,;\,t_0,T_i) \tag{4}$$

$$= 4\pi r_0 C_{se}\gamma_i\, D\,\varepsilon\,\{1 + \sqrt{\frac{K_i\, r_0^2}{\pi D\,(t-t_0)}}\, e^{-\lambda_i (t-t_0)} + \sqrt{\frac{\lambda_i\, K_i\, r_0^2}{D}}\, \mathrm{erf}\sqrt{\lambda_i\,(t-t_0)}\,\}\,\{h(t-t_0) - h(t-t_0-T_i)\,\}$$

where

K_i is the retardation coefficient of radionuclide i, dimensionless

D is the diffusion coefficient of radionuclide i, m^2/yr

λ_i is the decay constant of radionuclide i, yr^{-1}

r_0 is the radius of spherical waste form, m

C_{se} is the solubility of element e, g/m^3

γ_i is the inventory ratio of isotope to element, dimensionless

ϵ is the porosity of the diffusing medium, dimensionless

and $h(\tau)$ is the Heaviside step function.

● Inventory-limited model

For soluble species, for example, cesium, strontium, and iodine, one can only estimate the range of leach rates, since the solubilities of their usual compounds may be too large to limit their leaching. A lower limit to the leach rate would be the leach rate of the waste matrix, if they release congruently with the matrix. The congruently released species (or inventory-limited release) has the same fractional leach rate as the waste matrix, if both leach rates are normalized to the instantaneous inventory in the undissolved waste.

$$m_i(t\,;\,t_0,T_i)$$

$$= 4\pi r_0^2\, L_c\,(t-t_0)\,\frac{M_i^0\, e^{-\lambda_i t}}{M_c^0}\{h(t-t_0) - h(t-t_0-T_i)\} \tag{5}$$

where M_c^0 is the initial inventory of the cement waste matrix. The leach rate L_c of cement per unit surface area of the waste form is to be obtained experimentally.

● Solid-diffusion model

For extremely low solubility species such as Co-60, leaching is limited by the diffusive transport rate of the species in solid waste form.

$$m_i(t\,;\,t_0,T_i) = 16\, C_i^0\, D\, e^{-\lambda_i t} \sum_{n=1}^{\infty}\sum_{m=1}^{\infty} [\frac{4b}{(2n-1)^2\,\pi} + \frac{\pi}{b\,\alpha_m^2}]\, e^{-D(t-t_0)(\alpha_m^2+\beta_n^2)} \cdot\{h(t-t_0) - h(t-t_0-T_i)\,\} \tag{6}$$

where a is the radius of the cylindrical waste form, b is the half-height of the cylinder, C_i^0 is an initial concentration, α_m is the positive root of $J_0(a\alpha_m)=0$, $\beta_n=(2n-1)\pi/2b$.

2.4. Parametric Uncertainty and Sensitivity Analysis

For the parametric uncertainty and sensitivity analysis in the risk assessment of low-level radioactive waste repository, Latin hypercube sampling and rank correlation techniques[6] are applied for the parameter values shown in Table 1. Latin hypercube sampling technique is cost-effective for large computer programs because it generates smaller error to estimate output distribution even with smaller number of runs than Crude Monte Carlo technique. Rank correlation technique is good for generating dependence structure between samples of input parameters.

2.5. Exposure Dose Estimation

Among various exposure scenarios evaluated for the risk assessment, intruder scenario results in the worst exposure scenario. In this risk assessment, an imaginary town adjacent to the repository is considered as shown in Table 2. The assumed exposure pathways in the intruder scenario are:

a) extensive use of contaminated groundwater from an imaginary aquifer underlying the repository by the whole population of an imaginary town

Table 1. Parameter Values Used in the Analysis[7,8]

Parameter	Value	Distribution Type
Degradation time	0–300 years	lognormal
Corrosion rate	26.8–262 μm/yr	loguniform
Waste-form leach rate	10^{-3}–10^{-4} g/cm^2 day	uniform
Porosity	0.248–0.475	uniform
Drum thickness	Mean 1.6 mm Standard deviation 0.16 mm	normal
Mass of waste form per drum	Mean 3.1×10^5 g Standard deviation 6.2×10^4 g	normal
Diffusion coefficient in solid	10^{-10} cm^2/s	constant
Diffusion coefficient in liquid	315.36 cm^2/yr	constant

Table 2. Imaginary Town Adjacent to LLW Repository

Population		9500 persons
Area		52 km^2
Wells	–depth	100 m
	–pumping rate	200 m^3/yr
	–minimum distance btwn wells	500 m
	–number of wells	200
Water consumption rate by man		200 1/day
	–drinking	0.549 m^3/yr
Water consumption rate by cattle		29 m^3/yr

b) food grown on site.

The annual dose to an individual is calculated for the following biosphere pathways: [9,10]

a) drinking water
b) milk
c) beef
d) vegetables.

Groundwater flow model is shown conceptually in Figure 1.

3. Results

3.1. Radionuclide Release from Repository

The calculated annual release rates of 9 important radionuclides out of repository engineered barriers from 250,000 drums of nuclear power plant waste disposed of in an underground repository are shown in Figure 2.

Among various nuclides, long half-life nuclides such as Cs-137, Sr-90, and Tc-99 in fission products, Co-60, Ni-59, and Ni-63 in activation products, and Pu-241, Am-241, and Cm-244 in actinides are selected as the representative radionuclides[8] as shown in Table 3. The maximum initial radioactivity expected for the first low-level radioactive waste repository with the capacity of 250,000 drums is expected less than 134,000 Ci. About 50 % of the initial radioactive contents will result from Cs-137, Co-60, and Ni-63.

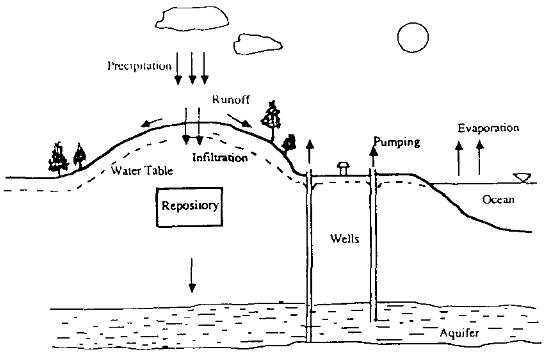

Fig. 1. Conceptual Groundwater Flow Model

3.2. Parametric Uncertainty and Sensitivity Analysis

In Figure 3, the annual release rate of Cs-137 which is the most important radionuclide for short-term to medium-term radiological effects of repository, is calculated for 30 sample runs. Due to conservatively chosen parameter values, some of the 30 sample runs result in very high annual release rate of Cs-137. The maximum release rate out of repository into geologic media among 30 sample runs is 170 Ci/yr, or 0.5% release of initial Cs-137 inventory per year.

In Figure 4, the rank correlation coefficients of some of the input parameters are shown. From the results of the sensitivity analysis, it is found that the degradation time of concrete structure and the corrosion rate of steel drum are strongly sensitive to overall uncertainty in few tens of years after repository closure and that in hundreds years after closure the leach rate of cement waste matrix is strongly sensitive to overall uncertainty in case of Cs-137.

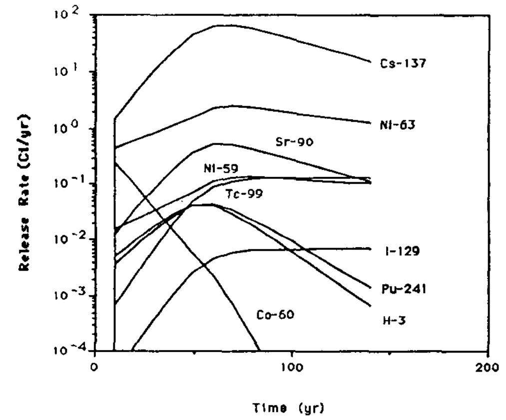

Fig. 2. Radionuclide Release from Repository (250,000 Drums)

Table 3. Radionuclide Inventory of LLW Repository[8] (250,000 drums)

Nuclide	Inventory [Ci]	Nuclide	Inventory [Ci]
H-3	158	Sb-125	258
C-14	169	I-129	0.63
Fe-55	23,065	Cs-134	11,808
Co-60	22,679	Cs-137	33,978
Ni-59	364	Eu-154	2.8
Ni-63	12,075	Pu-238	6.1
Sr-90	288	Pu-239	9.1
Nb-94	9.4	Pu-241	104
Tc-99	12	Am-241	11
Ru-106	1,811	Cm-244	2.6

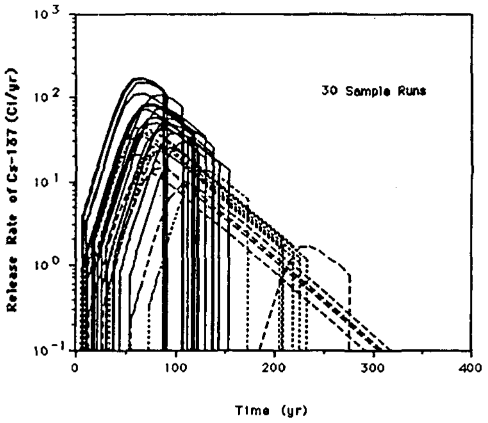

Fig. 3. Results of Parametric Uncertainty Analysis

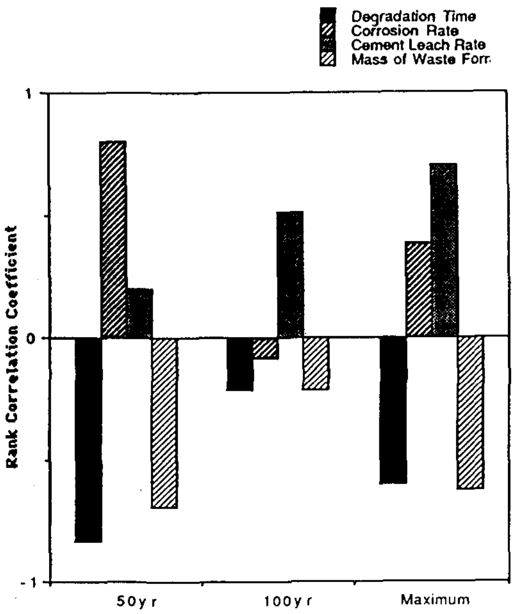

Fig. 4. Rank Correlation Coefficients of Important Input Parameters

3.3. Exposure Dose Estimation

Even for the worst-case scenario such as the intruder town just next to the repository site, extensive development of groundwater resources, no credit to natural barrier (no sorption retardation, no dispersion, and instantaneous transfer of released radioactivity from engineered barriers to biosphere), and very conservatively chosen parameters of the biosphere model, the maximum value of the annual dose to an individual among 30 sample runs is 3.96 mrem/year during the institutional control period (within 100 years) if aquifer size is assumed 100 times of annually pumped volume of groundwater and if this scenario is assumed to happen soon after repository closure. However, given the hypothetical nature of the scenario, the credit should be given to institutional control. Moreover, once the intrusion scenario occurs, reasonably conservative actions on the part of the intruders should be assumed to occur.

The exposure dose limits for the post-institutional control period, will be 1 mrem/year, which is *de minimis* level for individual specified by the ICRP. In reality, the worst-case scenario of the intruder town just next to the repository site may be meaningful only in the post-institutional control period, not during the institutional control period. A dose estimated to occur from the human intrusion within the repository boundary during the institutional control period should be viewed more as an illustration of what the dose would be without the institutional control. The result of this study suggests that the institutional control period of 100 years will be sufficient from the radiological safety point of view even for the worst-case scenario considered in this study.

According to a previous analysis[11], the maximum dose rate from low-level radioactive waste repository would be less than 1 mrem/year when natural barrier was taken into consideration. Therefore the interpretation of the results of this analysis requires some caution. In this study, we conservatively neglect the natural barrier to show that low-level radioactive waste repository would cause no serious harm to human even in unrealistic intruder scenario, and that the performance of far field needs to be proven for minimum requirements to ensure adequate repository performance.

4. Conclusions

A simplified safety assessment is carried out on rock-cavern type disposal for LLW. The results of the preliminary assessment show that Cs-137, Ni-63, and Sr-90 are dominant. From the results of uncertainty and sensitivity analysis of Cs-137 release, it is found that the degradation time of concrete structure and the corrosion rate of steel drum are very sensitive to overall uncertainty at early stage after repository closure and that the leach rate of cement waste matrix is sensitive to overall uncertainty on the long term. The results of the potential public health impacts associated with the low-level radioactive waste repository show that radiological dose to persons who might be exposed do not exceed the expected regulatory limits. It is also demonstrated that more attention should be given to near-field performance.

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