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The Analytical Radioactive Waste Repository Source Term REPS Model

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방사성폐기물 처분장 선원항 REPS 모델

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Abstract

The analytical repository source term (REPS) computer code is developed for the safety assessment of radioactive waste geologic repository. 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 this REPS code. For the validation of the radionuclide leach rates predicted by the REPS model, the calculated leach rates of Cs-137, Sr-85, and Co-60 are compared with two reported leaching test results. Cesium and strontium leach congruently, and the leaching test results of these species can be reproduced by the congruent leaching model included in the REPS model. In case of cobalt, the solid diffusion model is in good agreement with the leaching test results.

요 약

방사성폐기물 처분장의 안전성평가에 사용될 핵종유출 선원항 컴퓨터 코드 REPS를 개발하였다. 신뢰할만한 핵종별 침출율 예측을 위하여 REPS 코드에서는 콘크리트 구조물의 열하시간, 부식의 형태와 부식율, 드럼표면의 부식면적비, 그리고 핵종의 특성등이 고려되었다. 핵종유출 선원항 REPS 모델로부터 예측된 각 핵종별 침출율이 실제 실험결과와 어느정도 일치하는지를 알아보기 위하여 Cs-137, Sr-85, 그리고 Co-60 등을 선택하여 검증하였다. 세슘과 스트론튬은 조화용해 모형식을 사용하여 침출실험 데이타를 재현할 수 있었다. 이에 비해 침출이 느리게 일어나는 코발트의 경우 고화체내에서의 확산에 의한 침출 모형식이 적합함을 알 수 있었다.

I. Introduction

Most low- and intermediate-level radioactive wastes, such as liquid concentrates, from nuclear power plants are processed by cement solidi fication. The objective of the low- and intermediate-level radioactive waste repository source term model development is to develop a system model

capable of predicting radionuclide release rates from cemented waste drums disposed of in underground repository. Repository source-term model will be used for the performance assessment of geologic repository as described in Fig. 1. The system 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.

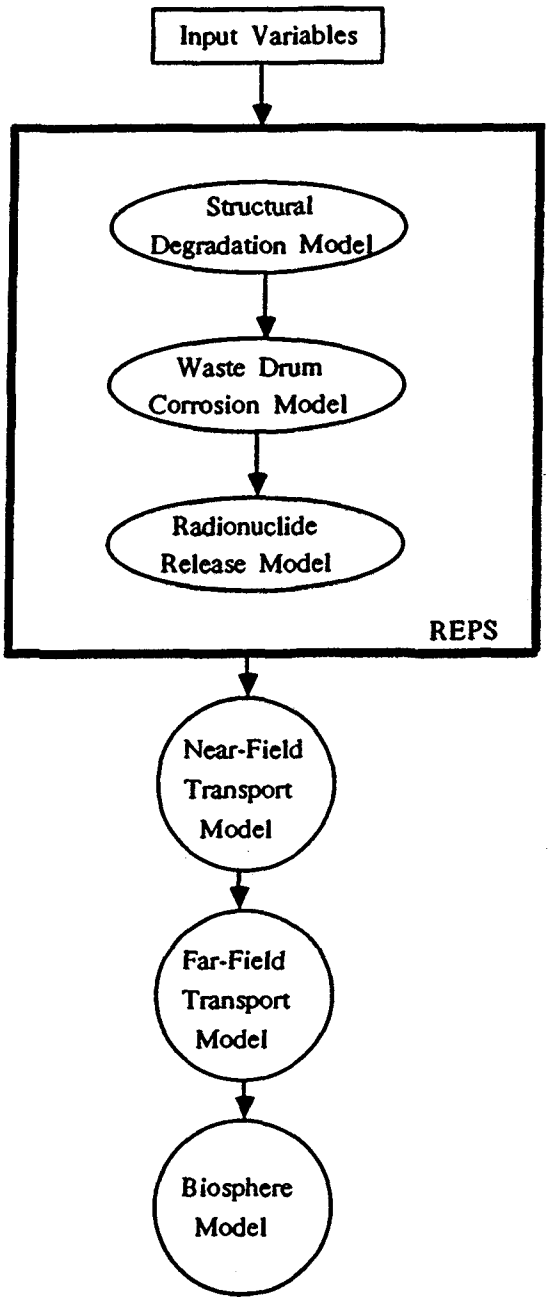

Fig. 1 Radioactive Waste Disposal Safety Assessment and REPS Model

II. Theoretical Consideration of REPS Computer Program

II-1. Structures of REPS

A primary objective of the REPS modeling is to make the solution procedure flexible enough to allow incorporation of new models to represent alternative disposal methods for high-level radioactive waste while retaining the basic procedures used for modeling low-level radioactive waste disposal. Therefore, the computer code is structured by a series of modules that represent physical processes. As shown in Fig.2, the current version of the repository source term (REPS) computer code consists of a main program and 14

No penetration of groundwater may occur while the concrete structure retains its integrity. The degradation of a reinforced concrete may proceed through the following possible processes : internal changes, chemical reactions with carbon dioxide and groundwater constituents, physical changes resulting from wet/dry or freeze/thaw cycles, corrosion of reinforcement, action of microorganisms, etc.[1] The predominant degradation mechanism could be changed with environmental characteristics of a specific disposal site. DEGRA subroutine determines the degradation time of concrete structure in the radioactive waste repository. Concrete structure's contribution to keeping radionuclides immobilized will be overlooked in this analysis for conservative and simple prediction.

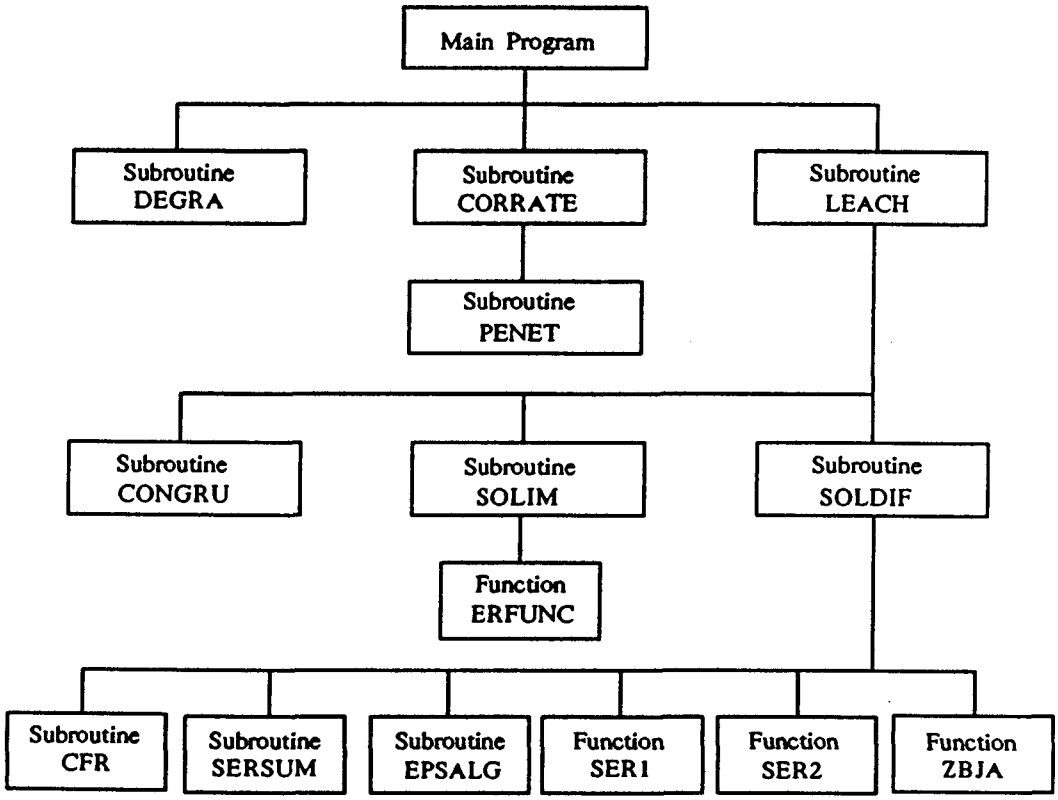

Fig. 2 Structures of the REPS Code

After degradation of concrete structure, groundwater may contact with the radioactive waste drums to corrode drum surface. CORRATE subroutine determines the corrosion rate of waste drum from the data base of conservatively estimated corrosion rates at various environments. PENET subroutine calculates container penetration time based on the determined corrosion rate and the thickness of radioactive waste drum. In this study, leaching of radionuclides from the waste form is assumed to be initiated by contact with groundwater that may occur associated with corrosion of the container. LEACH subroutine calculates the leach rates for various radionuclides. An extension of the concepts presented in Fig. 2 is shown in Fig.3.

II-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. Several mechanistic models for corrosion have been developed[2,3]. However, empirical models are preferred to mechanistic models to predict container corrosion rate because corrosion of steels is a complex process that depends on too many parameters such as groundwater Eh, pH, major ions present, type of steel, location of defects in the steel, surface impurities, etc. For this reason, corrosion model of REPS is empirical in nature and uses a corrosion data base. The gener-

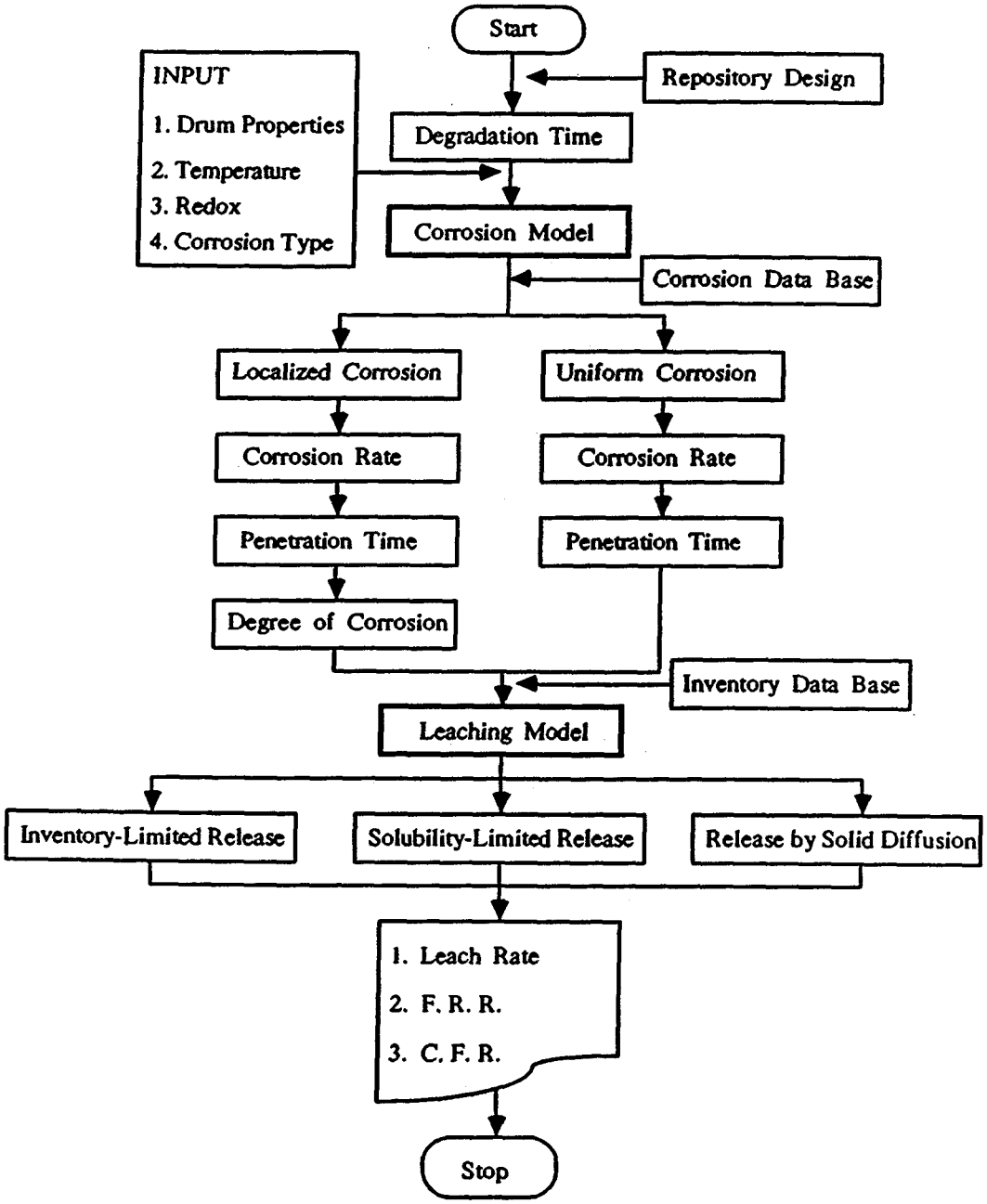

Fig. 3 Flow Diagram of the REPS Code

ıl form of corrosion depth equation is

$$d = k\,t^{n} \quad (1)$$

vhere d is the corrosion depth, k is a corrosion ate constant from the corrosion data base, t is time, and n is an empirical constant which gives the time dependence of corrosion depth. Modelers often set n equal to 1 for long-term corrosion depth prediction.

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 [4]

$$C_R(t) = \exp(\alpha + \beta t)/(1+\exp(\alpha + \beta t)) \quad (2)$$

where α and β are empirical constants determined by fitting experimental data to eq.(2).

II–3. Leaching Mechanisms

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 the following: (1) Solubility-limited release, (2) Congruent release, and (3) Solid diffusion controlled release.

(1) Solubility–Limited Release

The equation for the solubility–limited diffusive release of radionuclide i from a spherical waste form surface is

$$K_i \frac{\partial C_i(r,\tau)}{\partial \tau} = D \frac{1}{r^2} \frac{\partial}{\partial r} [r^2 \frac{\partial C_i(r,\tau)}{\partial r}] - \lambda_i K_i C_i(r,\tau); \quad r > r_0, \quad \tau > 0 \quad (3)$$

The initial condition is

$$C_i(r,0) = 0, \quad r > r_0 \quad (4)$$

The boundary conditions are

$$C_i(r_0,\tau) = C_{se}\gamma_i, \quad \tau \geq 0 \quad (5)$$

$$C_i(\infty,\tau) = 0, \quad \tau \geq 0 \quad (6)$$

where $C_i(r, \tau)$ is the concentration of radionuclide i in the groundwater, g/m^3

K_i is the retardation coefficient of species, dimensionless

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

λ_i is the decay constant of species, 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

and τ is the time after leaching begins, yr.

The desired solution of eqs. (3)–(6) is obtained using the Laplace transform and convolution theorem [5]

$$C_i(r,\tau) = \frac{C_{se}\gamma_i r_0}{2r} \{e^{(r-r_0)\sqrt{K_i\lambda_i/D}} \operatorname{erfc}[\frac{(r-r_0)}{2}\sqrt{K_i/(D\tau)} + \sqrt{\lambda_i \tau}] + e^{-(r-r_0)\sqrt{K_i\lambda_i/D}} \operatorname{erfc}[\frac{(r-r_0)}{2}\sqrt{K_i/(D\tau)} - \sqrt{\lambda_i\tau}]\}, \quad r > r_0, \ \tau > 0 \quad (7)$$

The quantity of concern is the total mass flux $\dot{m}_i(\tau)$ of species i from the waste form surface into the surrounding porous rock.

Since

$$\dot{m}_i(\tau) = 4\pi r_0^2 \{-\varepsilon D \frac{\partial C(r_0,\tau)}{\partial r}\}, \quad \tau > 0 \quad (8)$$

one obtains from eqs.(7) and (8)

$$\dot{m}_i(\tau) = 4\pi r_0 C_{se}\gamma_i D\varepsilon [1 + \sqrt{\frac{K_i r_0^2}{\pi D t}} e^{-\lambda_i\tau} + \sqrt{\frac{\lambda_i K_i r_0^2}{D}} \operatorname{erf}\sqrt{\lambda_i\tau}] \{h(\tau) - h(\tau - T_i)\}, \quad g/yr \ ; \ \tau > 0 \quad (9)$$

where $h(\tau)$ is the Heaviside step function and ε is the porosity of the diffusing medium.

The leach time T_i of radionuclide i is determined by the following equation

$$\frac{dM_i(\tau)}{d\tau} = -\dot{m}_i(\tau) - \lambda_i M_i(\tau), \quad 0 < \tau \leq T_i \quad (10)$$

with side conditions

$$M_i(0) = M_i^0 \quad (11)$$

$$M_i(T_i) = 0 \quad (12)$$

where M_i^0 is the initial inventory of radionuclide i per a waste package, $M_i(\tau)$ is the inventory at time τ in the waste form, and $\dot{m}_i(\tau)$ is the leach rate of radionuclide i at time τ.

(2) Inventory-Limited Release

For soluble species, for example, cesium, strontium, and iodine, one can only estimate the range of dissolution rates, since the solubilities of their usual compounds may be too large to limit their dissolution. 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 at any time τ after beginning of leaching as the waste matrix, if both leach rates are normalized to the instantaneous inventory in the undissolved waste. Then the leach rate $\dot{m}_i(\tau)$ of the congruently released species i at time τ, normalized to the inventory $M_i(\tau)$ of species i at time τ is the same as the leach rate $\dot{m}_c(\tau)$ of the cement waste matrix at time τ, normalized to the inventory $M_c(\tau)$ of the cement waste matrix at time τ:

$$\dot{m}_i(\tau) = 4\pi r_0^2 L_c(\tau) \frac{M_i(\tau)}{M_c(\tau)} [h(\tau) - h(\tau - T_c)] \qquad (13)$$

$L_c(\tau)$ in eq.(13) can be obtained by the following mechanistic model. As saturation of dissolved cement is approached, the net rate of solid-liquid reaction decreases to match the slow rate that the dissolved cement can diffuse from the surface liquid. Without assumptions as to which process is controlling, the time-dependent leach rate of cement from the surface of a waste solid into surrounding saturated porous media is obtained from the following diffusion equation using experimentally determined solid-liquid reaction rate as a boundary condition:

$$K_c \frac{\partial C_c}{\partial \tau} = D \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial C_c}{\partial \tau} \right), \quad \tau > 0, \; r_0 < r < \infty \qquad (14)$$

with initial condition

$$C_c(r,0) = 0, \quad r_0 < r < \infty \qquad (15)$$

As a boundary condition at the waste solid-liquid interface, the diffusive mass transfer rate of dissolved cement is equal to the dissolution rate by the solid-liquid reaction:

$$-\varepsilon D \frac{\partial C_c(r_0,\tau)}{\partial r} = j_0 \left[1 - \frac{C_c(r_0,\tau)}{C_{sc}} \right], \quad \tau > 0 \qquad (16)$$

where C_{sc} is the saturation concentration of the dissolved cement and j_0 is the experimental forward reaction rate of the species per unit external surface area. The remaining boundary condition is

$$C_c(\infty,\tau) = 0, \quad \tau > 0 \qquad (17)$$

The time-dependent leach rate of cement per unit surface area of the waste form is obtained by solving eqs.(13)-(16) following the solution method by Zavoshy et al.[6]:

$$L_c(\tau) = j_0 \frac{1 + R \exp\left[\frac{(1+R)^2 D\tau}{K_c r_0^2}\right] \operatorname{erfc} \sqrt{\frac{(1+R)^2 D\tau}{K_c r_0^2}}}{1+R} \qquad (18)$$

where the dimensionless flux ratio R is defined as

$$R = \frac{j_0 r_0}{\varepsilon D C_{sc}} \qquad (19)$$

(3) Release Limited by Solid Diffusion

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

$$\frac{\partial C_i}{\partial \tau} = D \left[\frac{\partial^2 C_i}{\partial r^2} + \frac{1}{r} \frac{\partial C_i}{\partial r} + \frac{\partial^2 C_i}{\partial z^2} \right] - \lambda_i C_i, \quad 0 \le r \le a, \; -b \le z \le b \qquad (20)$$

where a is the radius of the cylindrical waste form, and b is half-height of the cylinder. The initial condition is

$$C_i(r,z,0) = C_i^0 \qquad (21)$$

where C_i^0 is an initial concentration. The boundary conditions are

$$C_i(a,z,\tau) = 0, \quad \tau > 0 \tag{22}$$

$$C_i(r,\pm b,\tau) = 0, \quad \tau > 0 \tag{23}$$

The solution of eqs. (20)–(23) was obtained by Nestor[7] :

$$C_i = \frac{8C_i^0 e^{-\lambda_i \tau}}{\pi a} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{(-1)^n J_0(r\alpha_m)}{(2n-1)\alpha_m J_1(a\alpha_m)} \cos\beta_n z \exp[-D\tau(\alpha_m^2 + \beta_n^2)] \tag{24}$$

where

J_0 is the zero-order Bessel function,

J_1 is the first-order Bessel function,

α_m is the positive root of $J_0(a\alpha_m)=0$,

and $\beta_n=(2n-1)\pi/2b$.

Finally the time-dependent diffusion-controlled leach rate of species i from the surface of a cylindrical waste form is

$$\dot{m}_i(\tau) = 16C_i^0 D e^{-\lambda_i \tau} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \left[\frac{4b}{(2n-1)^2\pi} + \frac{X_0 \pi}{b\alpha_m^2}\right] e^{-D\tau(\alpha_m^2+\beta_n^2)} \cdot \{h(\tau) - h(\tau - T_i)\} \tag{25}$$

II-4. Leach Model

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

$$f_i(t) = \frac{\dot{m}_i(t\,;\,t_0, T_i)\, C_R(t)}{M_i^0} \tag{26}$$

where $\dot{m}_i(t\,;\,t_0, T_i)$ is the leach rate of radionuclide i from a waste format time t after emplacement, t_0 is the container penetration time or the starting time of leaching after emplacement, T_i is the leach time or the duration of leaching for radionuclide i, and $C_R(t)$ is the exposed surface area ratio given in eq. (2).

The difference between the time after emplacement t and the time after beginning of leaching is shown in Fig. 4.

$\dot{m}_i(t\,;t_0,T_i)$ has a non-zero value for $t_0 \le t \le T_i + t_0$.

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

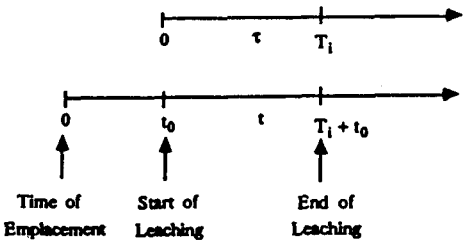

Fig. 4 Comparison of t and τ

(1) Solubility–limited model

From eq.(9)

$$\dot{m}_i(t\,;t_0,T_i) = 4\pi r_0 C_e \gamma_i D\varepsilon \left\{1 + \sqrt{\frac{K_i r_0^2}{\pi D(t-t_0)}}\right\} e^{-\lambda_i(t-t_0)} + \sqrt{\frac{\lambda_i K_i r_0^2}{D}}\, \mathrm{erf}\sqrt{\lambda_i(t-t_0)} \} \{h(t-t_0) - h(t-t_0-T_i)\} \tag{27}$$

(2) Inventory-limited model

From eq.(13)

$$\dot{m}_i(t\,;t_0,T_i) = 4\pi r_0^2 L_c(t-t_0) \frac{M_i^0 e^{-\lambda_i t}}{M_c^0} \{h(t-t_0) - h(t-t_0-T_i)\} \tag{28}$$

where the leach rate of cement per unit area of waste form $L_c(t)$ is given in eq.(18).

(3) Solid-diffusion model

From eq.(25)

$$\dot{m}_i(t\,;t_0,T_i) = 16C_i^0 D e^{-\lambda_i t} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \left[\frac{4b}{(2n-1)^2\pi} + \frac{\pi}{b\alpha_m^2}\right] e^{-D(t-t_0)(\alpha_m^2+\beta_n^2)} \cdot \{h(t-t_0) - h(t-t_0-T_i)\} \tag{29}$$

The cumulative fractional release of radionuclide i from a waste drum is

$$F_i(t) = \int_0^t f_i(t')\,dt' \tag{30}$$

III. Results and Discussion

III-1. Calculation of Radionuclide Leach Rates

As an illustration of the REPS model, the calculated leach rates of Cs-137, Sr-85, and Co-60 are compared with two reported leaching test results[8,9]. Leaching experiments of Cs-137 and Sr-85 by Moore[8] used small simulated cement waste forms of 5.7 g. However, leaching experiments of Co-60 by Croney[9] used both real cement waste forms of 56 cm diameter and 76 cm height (55 gallon drum) and small cylindrical samples of 5 cm diameter and 10 cm height produced by the solidification of PWR boric acid waste.

The inventory-limited leaching model in the REPS code is included to predict the soluble radionuclide leaching. The leaching of soluble Cs-137 and Sr-85 is assumed to be controlled by the leaching of the cement waste form matrix. Because the composition of cement waste form is complex, C_{sc}=1,310 g/m^3[10] and j_0=100 g/m^2 day [11], which are conservative values for solubility and forward reaction rate, are chosen for the calculation of cement leach rate with eq.(18) to get an upper bound estimate of cement waste form leach rate.

The cumulative fractional release of Cs-137 as a function of time calculated from the REPS code is shown in Fig.5. For the comparison, a result of Cs-137 leaching experiment by Moore[8] is also shown in Fig.5. The predicted cumulative fractional release is in good agreement with the leaching test result.

Similarly, the time-dependent cumulative fractional release of Sr-85 obtained by Moore[8] is in good agreement with the calculated cumulative fractional release by the congruent release model of the REPS code as shown in Fig.6.

Unlike cesium and strontium, cobalt is almost insoluble in water. To predict long-term leaching behavior of Co-60, solid diffusion model of the REPS code is used. The cumulative fractional release of Co-60 obtained by Croney[9] for small cylindrical waste form is compared with the calculated time-dependent cumulative fractional release by the REPS code in Fig.7. The long-term bounding estimation of the slow leach rate of insoluble Co-60 can be obtained by the REPS code. For the case of full size waste form with a smaller surface-to-volume ratio, the calculated leach rate by the REPS code is also in good agreement with

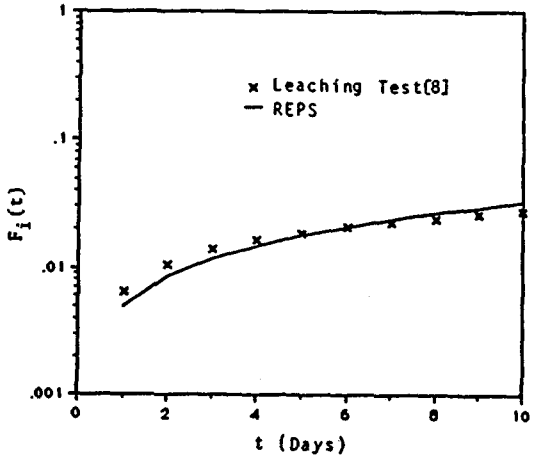

Fig. 5 Cumulative Fractional Release of Cs-137 from a Cement Waste Form

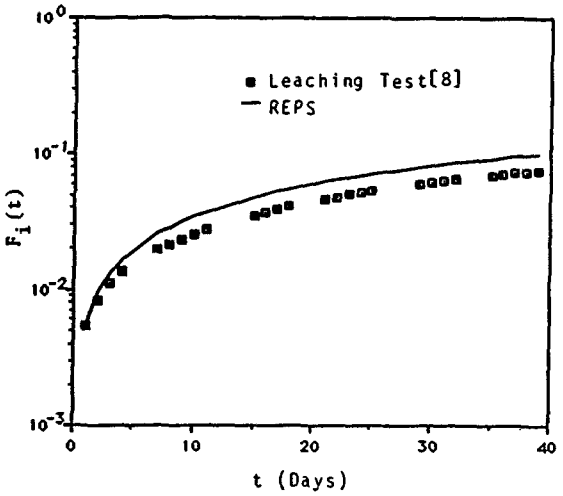

Fig. 6 Cumulative Fractional Release of Sr-85 from a Cement Waste Form

the lower leach rate observed by Croney[9].

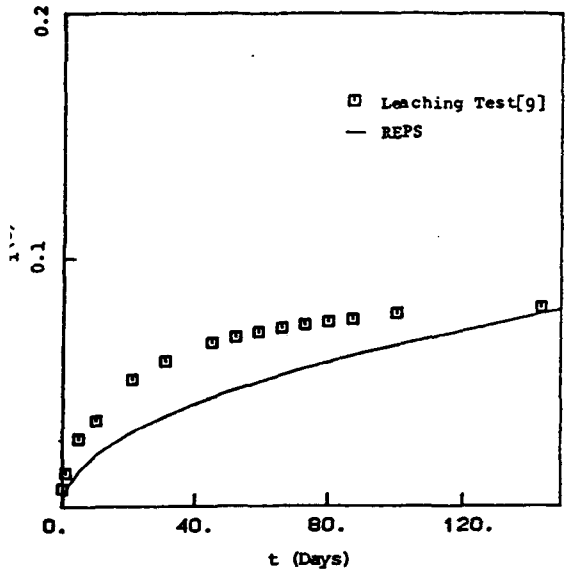

Fig. 7 Cumulative Fractional Release of Co-60 from a Cement Waste Form

III-2. Effect of Corrosion

Three different fractional release rates of Cs-137 for three waste container corrosion scenarios are shown in Fig.8, respectively. The first scenario is groundwater penetration by localized corrosion on carbon steel drum surface. A corrosion rate of 262 μm/yr is chosen from the corrosion data base of the REPS code for this illustration. The calculated penetration time and leach time are 4.5 years after emplacement and 45.9 years after leaching starts, respectively. The second scenario is groundwater penetration by uniform corrosion. The assumed corrosion rate is 26.8 μm/yr. For this case the calculated penetration time is 44.8 years after emplacement. The last case is the gradual increase of corroded surface area on waste drum. $\alpha = -5.3$ and $\beta = 0.12$ in eq.(2) are assumed for the illustration[4]. The calculated leach time is 90.1 years.

The first scenario results in the most conservatively high fractional release rate of Cs-137 because of the assumption that all the surface of drum is exposed to groundwater as soon as the first corrosion pit penetrate the drum. Because of the relatively lower corrosion rate of uniform corrosion, the release of Cs-137 for the second scenario is delayed and shows a lower maximum release rate. The last scenario may result in a probable and less conservative estimation of radionuclide release from waste drums in a geologic repository.

IV. Conclusions

A low-x_0 and intermediate-level radioactive waste repository source term code is developed and the preliminary calculation shows that the REPS code can estimate reasonably the leaching behaviors of Co, Cs, and Sr. The effect of exposed surface area ratio on the release of radionuclides from the waste drums can not be neglected. Therefore, details on the container corrosion, especially localized corrosion, should be studied. Leaching of other radionuclides including tritium, C-14, plutonium, americium, and iodine will be tested by three leaching models of the REPS code.

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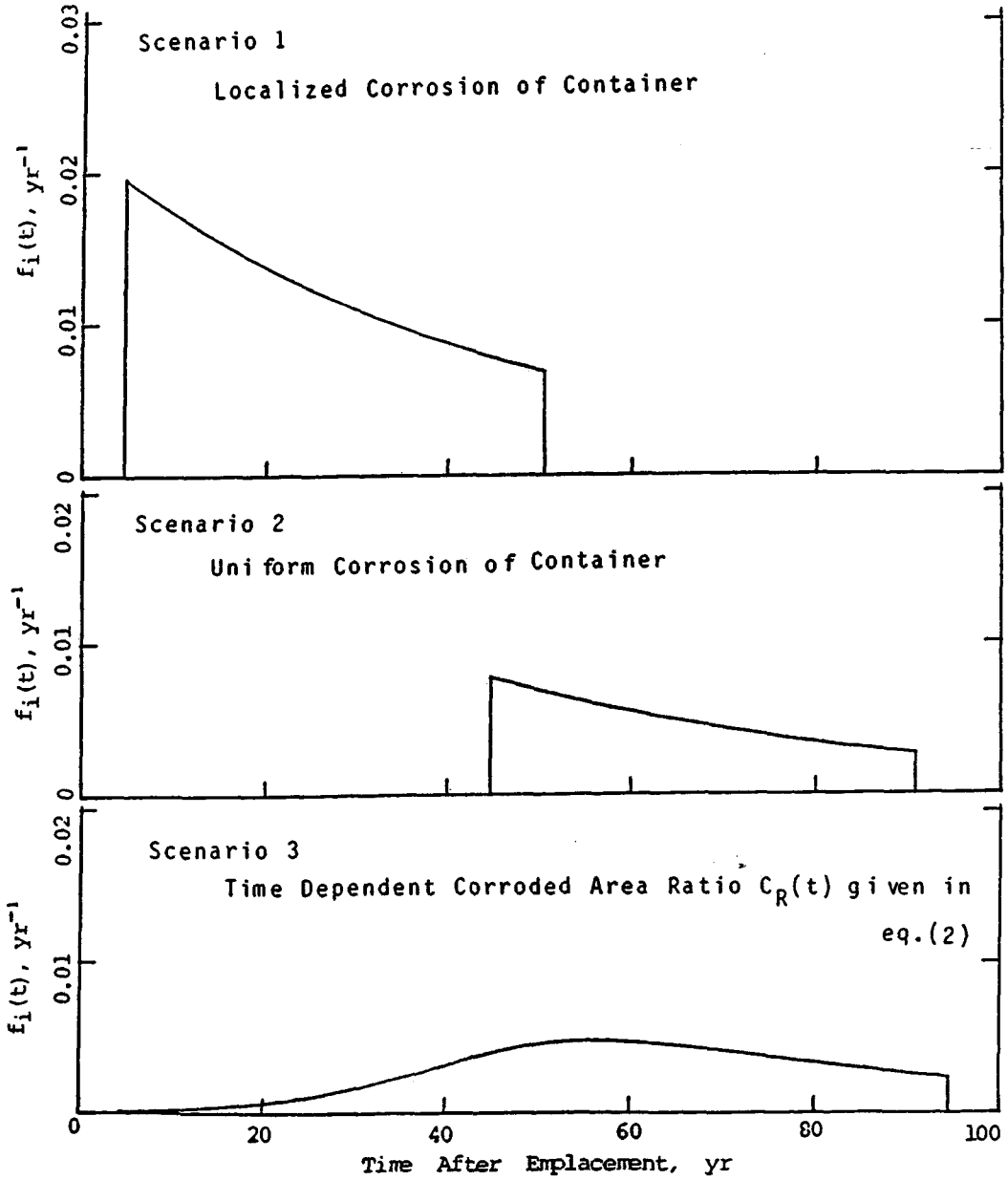

Fig. 8 Fractional Release Rates of Cs-137 Based on Three Different Waste Container Corrosion Scenarios

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