

Near-Field Transport of Radionuclide Decay Chains

Chul-Hyung Kang

Korea Atomic Energy Research Institute

(Received January 17, 1994)

방사성 핵종 붕괴 사슬의 Near-Field 이동

강 철 형

한국원자력연구소

(1994. 1. 17 접수)

Abstract

Much attention has been given to predict the near-field mass transfer of a single radioactive species from a waste solid into surrounding porous medium. But only limited considerations have been given to predict the coupled mass transfer of species with a radioactive decay chain. In this study we present an analysis assuming that the members of a decay chain dissolve congruently with a solubility-limited matrix. We give general, non-recursive analytic solutions for the transport of a radioactive decay chain in a finite porous medium when nuclides are released congruently with the matrix. As an illustration we consider the decay chain $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ from spent fuel. These solutions may be useful and potentially important in performance assessment of radioactive waste repositories.

요 약

폐기물 고화체로부터 주위의 Near-field로의 단일 핵종의 이동에는 많은 연구가 수행되고 있으나, 방사성 핵종 사슬의 이동에 관한 연구는 매우 제한되어 있다. 본 논문에서는 조화 유출되는 방사성 핵종 사슬의 이동을 분석하고, 유한 크기의 다공성 매질에서의 일반적인 비순환 해석해를 제시하였다. 또한 이해를 사용후 핵연료에서 가장 중요한 핵종사슬인 $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ 에 적용하여 보았다. 본 연구는 방사성 폐기물의 처분장 성능평가에 유용하고 중요하게 사용될 것이다.

1. Introduction

In radioactive waste and mixed waste studies, much attention has been given to predict the transport of a single radioactive species from the waste form into surrounding porous medium, But only limited considerations have been given to

predict the coupled transport of species in a radioactive decay chain. Failure to account for nuclides generated during transport may underestimate releases to the biosphere.

Although many solutions and computer codes for the transport of radioactive chains exist, most of them have limitations. Some use approximation

approaches. Some are limited by the number of chain members. Others give a recursive solution. Chambré et al. [1] obtained the general, non-recursive analytic solution for the transport of radioactive chains of arbitrary length. Lung et al. [2] implemented the general solution in finite and semi-infinite porous media for time-dependent mass transfer assuming that each member of the decay chain is at a specified constant concentration at the waste surface and for band release at the waste surface. In the analysis of the concentration boundary condition, however, there were unresolved problems, e.g., the interior maximum in the concentration profile which exceeds the surface concentration prescribed at the solubility limit, or the negative mass flux at the waste surface. These solutions may be useful for many situations with radioactive and mixed wastes where a constant concentration, such as the solubility, can be specified. However, for radioactive species dissolving from a solid waste form, as in high-level radioactive waste or spent fuels, a more sophisticated analysis is needed. In this study, we solved the equations, assuming, more realistic, that the members of arbitrary length of a decay chain dissolve congruently with solubility limited matrix.

2. Theoretical Analysis

Congruent dissolution of two species means that the ratio of the time-dependent mass rates of dissolution of those species equals the ratio of the time dependent concentrations of those species in the dissolving solid. Congruent dissolution is expected if there is no preferential leaching of any constituent in the waste solid. For example in a reducing environment the UO_2 matrix of spent fuel appears to dissolve congruently with the soluble constituents of spent fuel. However, if any constituent is of sufficiently low solubility, the species may not be able to dissolve all as it is released from the solid matrix by uranium dissolution. The dissolution of the low-

solubility species is not congruent with the dissolution of the uranium, although the release of the species and uranium from the UO_2 matrix solid may be congruent. The released low-solubility species can form a precipitate at the waste surface.

Here it is assumed that a matrix species whose dissolution is controlled by solubility, and other species dissolve congruently with the matrix. In light of the introduction, the "matrix" for this purpose could be uranium dioxide in spent nuclear fuel in a reducing environment, or it could be the precipitate of ^{238}U , ^{234}U , and other uranium isotopes that will form on the surface of spent fuel in an oxidizing environment or on glass waste. In this case we have to deal separately with the matrix species and the chain members.

The governing equation for the matrix species is

$$K_m \frac{\partial N_m}{\partial t} + v \frac{\partial N_m}{\partial z} + \lambda_m K_m N_m = D \frac{\partial^2 N_m}{\partial z^2}, \quad 0 < z < L, \quad t > 0 \quad (1)$$

which is to be solved for the concentration of the matrix species, $N_m(z, t)$. Here subscript m means the matrix species, D is the dispersion coefficient, K_m the retardation coefficient, λ_m the decay constant, and v the groundwater pore velocity.

With the initial condition

$$N_m(z, 0) = 0, \quad 0 < z < L \quad (2)$$

and the boundary conditions

$$N_m(0, t) = c_s, \quad t \geq 0 \quad (3)$$

$$-D \epsilon \frac{\partial N_m}{\partial z} + v \epsilon N_m = h N_m \quad \text{for } z = L, \quad t > 0. \quad (4)$$

where c_s is the solubility limited concentration of the matrix species, and h is the mass transfer coefficient describing the mass transport at $z=L$, and is assumed to be constant. From a previous analysis [3] one can estimate the time necessary for the transient mass transfer coefficient to reach a steady state value. For example, for cylindrical waste form the criterion for the time necessary to establish the steady state mass transfer coefficient is given by $Ut/Kr_0 \approx 1.2$. Here

U is the ground water pore velocity, r_0 is the radius of the waste form, and K is the retardation coefficient. For $U = 1$ m/yr and $r_0 = 25$ cm, the time to reach the steady state varies from 3 years to 300 years, as the retardation coefficient varies from $K=10$ to $K=1000$. This time period is much smaller than the time period in which we are interested. Thus one can assume reasonably the mass transfer coefficient to be constant for all time for this range of ground water velocity. For very low ground water velocity or very high retardation coefficient, one may use different mass transfer coefficients at early times and later times. This early time mass transfer coefficient may be obtained by averaging the transient mass transfer coefficient over time.

The governing equations for the members of the decay chain are

$$K_1 \frac{\partial N_1}{\partial t} + v \frac{\partial N_1}{\partial z} + \lambda_1 K_1 N_1 = D \frac{\partial^2 N_1}{\partial z^2}$$

$$K_2 \frac{\partial N_2}{\partial t} + v \frac{\partial N_2}{\partial z} + \lambda_2 K_2 N_2 = D \frac{\partial^2 N_2}{\partial z^2} + \lambda_1 K_1 N_1 \quad (5)$$

$$\dots\dots\dots$$

$$K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} + \lambda_i K_i N_i = D \frac{\partial^2 N_i}{\partial z^2} + \lambda_{i-1} K_{i-1} N_{i-1}$$

where $N_i \equiv N_i(z, t)$ is the concentration of the i^{th} member. The functions $N_i(z, t)$, $i=1, 2, \dots$ are subject to the initial conditions

$$N_i(z, 0) = 0, \quad 0 < z < L \quad (6)$$

and the type III boundary condition at the waste surface

$$-D\epsilon \frac{\partial N_i}{\partial z} + v\epsilon N_i = \dot{M}_i(t) \quad \text{for } z = 0, \quad t > 0 \quad (7)$$

where ϵ is the porosity.

The boundary condition at $z=L$ for the i^{th} member is

$$-D\epsilon \frac{\partial N_i}{\partial z} + v\epsilon N_i = hN_i \quad \text{for } z = L, \quad t > 0. \quad (8)$$

$\dot{M}_i(t)$ is determined from the definition of the congruent release

$$\frac{\dot{M}_m(t)}{\dot{M}_i(t)} = \frac{\dot{M}_i(t)}{\dot{M}_i(t)}, \quad t \geq 0 \quad (9)$$

or

$$\dot{M}_i(t) = \dot{M}_m(t) \frac{M_i(t)}{M_m(t)}, \quad t \geq 0.$$

where $M_m(t)$ and $\dot{M}_m(t)$ are the inventory and the mass flux of the matrix species, respectively, and $M_i(t)$ and $\dot{M}_i(t)$ are those of the i^{th} member.

To solve this system one proceeds as follows.

1. Solve the governing equation, (1), with side conditions, (2), (3) and (4), and compute the mass flux of the matrix species $\dot{M}_m(t)$ at the inner interface.
2. From equation (9) compute $\dot{M}_i(t)$.
3. Solve the governing equations, (5) with appropriate side conditions, (6), (7) and (8), for the chain members.

To obtain the concentration of the matrix species, $N_m(z, t)$, let

$$N_m(z, t) = U(z) + W(z, t) \quad (10)$$

where $U(z)$ satisfy

$$v \frac{dU}{dz} + \lambda_m K_m U = D \frac{d^2 U}{dz^2}, \quad 0 < z < L \quad (11)$$

with side conditions

$$U(0) = c_s \quad (12)$$

$$-D\epsilon \frac{dU}{dz} + v\epsilon U = hU \quad \text{for } z = L. \quad (13)$$

The solution $U(z)$ is given by

$$U(z) = e^{z v / 2D} u(z), \quad 0 \leq z \leq L \quad (14)$$

where

$$u(z) = (ae^{-q_m z} + be^{q_m z}) \quad (15)$$

$$a = \frac{c_s(\alpha_2 + q_m)e^{q_m L}}{(\alpha_2 + q_m)e^{q_m L} - (\alpha_2 - q_m)e^{-q_m L}} \quad (16)$$

$$b = \frac{-c_s(\alpha_2 - q_m)e^{-q_m L}}{(\alpha_2 + q_m)e^{q_m L} - (\alpha_2 - q_m)e^{-q_m L}} \quad (17)$$

$$q_m^2 = \left[\frac{K_m \lambda_m}{D} + \left(\frac{v}{2D} \right)^2 \right] \quad (18)$$

$$\alpha_2 = \frac{h - v\epsilon/2}{D\epsilon} \quad (19)$$

Now $W(z, t)$ must satisfy

$$K_m \frac{\partial W}{\partial t} + v \frac{\partial W}{\partial z} + \lambda_m K_m W = D \frac{\partial^2 W}{\partial z^2}, \quad 0 < z < L, \quad t > 0 \quad (20)$$

with the side conditions

$$W(z, 0) = -U(z), \quad 0 < z < L \quad (21)$$

$$W(0, t) = 0, \quad t > 0 \quad (22)$$

$$-D \epsilon \frac{\partial W}{\partial z} + v \epsilon W = h W \quad \text{for } z = L, \quad t > 0. \quad (23)$$

The solution $W(z, t)$ is given by

$$W(z, t) = e^{z v / 2 D} \sum_{i=1}^{\infty} A_i \sin(\beta_i z) e^{-\lambda_{im} t}, \quad 0 \leq z \leq L, \quad t \geq 0 \quad (24)$$

where

$$\lambda_{im} = \frac{D}{K_m} (\beta_i^2 + q_m^2) \quad (25)$$

$$A_i = \frac{-1}{\|\phi_i\|^2} \int_0^L \phi_i(z) u(z) dz \quad (26)$$

$$\|\phi_i\|^2 = \int_0^L \phi_i^2(z) dz. \quad (27)$$

The functions $\phi_i(z)$ are given by

$$\phi_i(z) = \sin(\beta_i z) \quad (28)$$

with eigenvalues β_i given by

$$\tan(\beta_i L) = -\frac{\beta_i}{\alpha_2}. \quad (29)$$

Putting them all together

$$N_m(z, t) = e^{z v / 2 D} [u(z) + \sum_{i=1}^{\infty} A_i \sin(\beta_i z) e^{-\lambda_{im} t}], \quad 0 \leq z \leq L, \quad t \geq 0 \quad (30)$$

The release rate $\dot{M}_m(t)$ is given by

$$\begin{aligned} \dot{M}_m(t) &= (-D \epsilon \frac{\partial N_m}{\partial z} + v \epsilon N_m) |_{z=0} \\ &= \frac{c_s v}{2} - D \epsilon e^{z v / 2 D} [u'(0) + \sum_{i=1}^{\infty} 2 A_i \beta_i e^{-\lambda_{im} t}] \end{aligned}$$

$$= \epsilon \psi(t), \quad t \geq 0. \quad (31)$$

where

$$\psi(t) = \frac{c_s v}{2} - D \epsilon e^{z v / 2 D} [u'(0) + \sum_{i=1}^{\infty} 2 A_i \beta_i e^{-\lambda_{im} t}]$$

On substitution of (31) in to (9) one obtains

$$\dot{M}_i(t) = \epsilon \psi(t) \frac{M_i(t)}{M_m(t)}, \quad t \geq 0. \quad (32)$$

Because the dissolution rate of the matrix is usually so low, one can neglect the mass loss of the matrix and decay members due to the dissolution. Then the inventory can be written

$$M_m(t) \simeq M_m^0 e^{-\lambda_m t}, \quad t \geq 0 \quad (33)$$

$$M_i(t) \simeq \sum_{j=1}^i B_{ij} e^{-\lambda_j t}, \quad t \geq 0. \quad (34)$$

Here M_m^0 is the initial inventory of the matrix, and B_{ij} is the coefficient of the Bateman equation

$$B_{ij} = \sum_{k=1}^j \left(\frac{M_k^0}{\lambda_i} \prod_{r=k}^i \lambda_r \right) / \prod_{\substack{l=k \\ l \neq j}}^i (\lambda_l - \lambda_j) \quad (35)$$

where M_k^0 is the initial inventory of the k^{th} member.

Substitution of (33) and (34) into (32), then yields the flux of the radionuclide

$$\dot{M}_i(t) = \epsilon \sum_{j=1}^i \frac{B_{ij}}{M_m^0} e^{-(\lambda_j - \lambda_m) t} \psi(t), \quad t \geq 0. \quad (36)$$

To avoid mathematical and numerical difficulties in solving the field concentration of chain members, we divided the solution into two modules: (1) a module to compute the boundary concentration with congruent release model, and (2) a module to compute the field concentration using the boundary concentration.

First solve the boundary concentrations of the chain members, $N_i(0, t)$, then

$$\begin{aligned} N_i(0, t) &= \frac{2}{M_m^0 K_i} \left[\sum_{k=1}^i B_{ik} \right. \\ &\quad \left. + \sum_{l=1}^{\infty} \frac{\beta_l^2}{(\beta_l^2 + \alpha_1^2)(L + \frac{\alpha_2}{\beta_l^2 + \alpha_2^2}) + \alpha_1} \operatorname{conv}(p; k, i) \right] dp + \end{aligned}$$

$$+ \sum_{j=1}^{i-1} \prod_{m=j}^{i-1} \lambda_m \sum_{n=j}^i \sum_{l=1}^{\infty} \frac{1}{\prod_{r=n}^i (\Gamma_{nr} \beta_l^2 + \gamma_{nr})} \frac{\beta_l^2}{(\beta_l^2 + \alpha_1^2)(L + \frac{\alpha_2}{\beta_l^2 + \alpha_1^2}) + \alpha_1} \cdot conv(p; k, n) dp], \quad t \geq 0 \quad (37)$$

where

$$\begin{aligned} conv(p; i, n) &= [\psi(t) e^{-(\lambda_i - \lambda_m)t}] * e^{-\delta_n t} \\ &= \int_0^t \psi(\tau) e^{-(\lambda_i - \lambda_m)\tau} e^{-\delta_n(t-\tau)} d\tau \\ &= \frac{v}{2} \frac{1}{\lambda_i - \lambda_m - \delta_n} (e^{-\delta_n t} - e^{-(\lambda_i - \lambda_m)t}) - \\ &\quad \frac{D^2}{K_m} \sum_{r=1}^{\infty} \frac{2}{L + l_r} \frac{\beta_r^2}{\delta_m} \times \\ &\quad \times \left[\frac{1}{\lambda_i - \lambda_m - \delta_n} (e^{-\delta_n t} - e^{-(\lambda_i - \lambda_m)t}) - \right. \\ &\quad \left. - \frac{1}{\lambda_i - \lambda_m - \delta_n + \delta_m} (e^{-\delta_n t} - e^{-(\lambda_i - \lambda_m + \delta_m)t}) \right], \quad t \geq 0. \end{aligned} \quad (38)$$

$$\alpha_1 = v/2D \quad (39)$$

$$\alpha_2 = \frac{h - v\epsilon/2}{D\epsilon} \quad (40)$$

$$q_n^2 = \left[\frac{K_n \lambda_n}{D} + \alpha_2^2 \right] \quad (41)$$

$$\delta_n = \frac{D}{K_n} (\beta_l^2 + q_n^2) \quad (42)$$

$$\Gamma_{nr} = D \left(\frac{1}{K_r} - \frac{1}{K_n} \right) \quad (43)$$

$$\gamma_{nr} = [(\lambda_r - \lambda_n) - \alpha_1^2 \Gamma_{nr}] \quad (44)$$

$$l_i = \frac{\alpha_2}{\beta_l^2 + \alpha_2^2} \quad (45)$$

with eigen values β_l given by

$$\tan \beta_l L = \frac{\beta_l(\alpha_1 + \alpha_2)}{\beta_l^2 - \alpha_1 \alpha_2} \quad (46)$$

Next, we solve the system with arbitrary concentration boundary condition, i. e., $N_i(0, t) = f_i(t)$.

Then the solution becomes

$$N_i(z, t) = e^{vz/2D} \frac{D}{K_i} \left[\sum_{l=1}^{\infty} \left(\frac{2}{L + l_i} \right) \beta_l \sin \beta_l z f_i(t) * e^{-\delta_i t} + \right.$$

$$\left. + \sum_{j=1}^{i-1} \prod_{m=j}^{i-1} \lambda_m \sum_{n=j}^i \sum_{l=1}^{\infty} \left(\frac{2}{L + l_i} \right) \frac{\beta_l \sin \beta_l z}{\prod_{r=n}^i (\Gamma_{nr} \beta_l^2 + \gamma_{nr})} f_j(t) * e^{-\delta_n t} \right], \quad z > 0, \quad t > 0, \quad i = 1, 2, \dots \quad (47)$$

The details of this analysis are given by Chambré et al. [2] and Kang [4]. For the computation of the infinite summations in (37) and (47), one may adopt the $e_m(S_n)$ transformation, which was developed by Shanks [5].

3. Numerical Illustrations

We use the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ chain in a spent fuel matrix of UO_2 to illustrate the solution. This is an important chain in the safety assessment of radioactive waste repositories. The inner layer we consider might consist of backfill around a radioactive waste package, or a damaged rock region. The values used are here for a backfill around a waste package in a wet-rock repository: inner layer thickness $L = 0.30$ m, mass transfer coefficient $h = 10^{-4}$ m/yr, groundwater velocity $v = 0$ m/yr, diffusion coefficient $D = 10^{-5}$ cm^2/s and porosity of the inner layer $\epsilon = 0.3$ [2, 6]. Table 1 shows data on the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ chain.

Figure 1 shows the normalized flux of the matrix species and each member of the decay chain, $^{234}\text{U} \rightarrow$

Table 1. Data Used in Illustrations

Nuclide	^{238}U	^{234}U	^{230}Th	^{226}Ra
Ki	120	120	1500	300
$T_{1/2}$, yr	4.5×10^5	7.7×10^4	1.6×10^3	
M_m^0 or M_i^0 , g	1.74×10^6	338	0	0

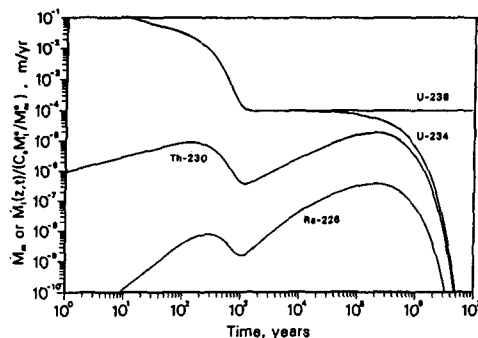


Fig. 1. Normalized Matrix Species and Radionuclides Mass-Fluxes at the Waste-Backfill Interface as a Function time.

$^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ at the waste surface. The flux of the matrix species decreases as time increases and ^{238}U fills up in the backfill region. At about $t \approx 300$ years the backfill is almost filled up, and the flux decreases rapidly. The ^{238}U reaches steady state about $t \approx 1000$ years. The trend of the flux of ^{234}U is almost the same as that of the matrix species until the decay effect of ^{234}U becomes important at $t \approx 10^5$ years. The fluxes of ^{230}Th and ^{226}Ra have dips at $t \approx 1000$ years. These dips come from the fact that the flux of the each member depends on the inventory and the flux of the matrix species, see (9). In early times the flux of the matrix species decreases, while the inventories of ^{230}Th and ^{226}Ra increase due to the decay of the ^{234}U . The increasing rates of these inventories exceed the decreasing rate of the flux of the matrix species. Thus the fluxes of these two daughters increase. However, for the time period, $300 < t < 1000$ years the flux of the matrix species decreases rapidly, thus the fluxes of ^{230}Th and ^{226}Ra decrease even though their inventories still increase. After this time period the fluxes increase until the decay effect of ^{234}U becomes important.

Figure 2 shows normalized concentrations as a function of time at the interface of the two layers, $z=L$, shown as a solid curve. For comparison, the concentration at the waste surface, $z=0$, is also shown. The concentration at the layer interface is

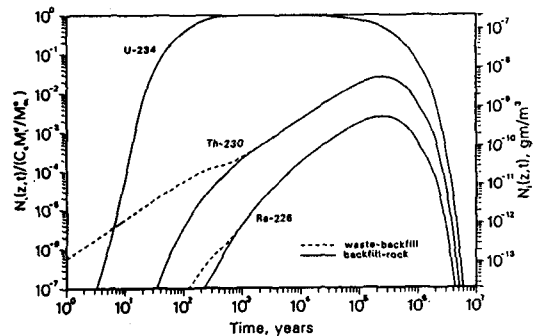


Fig. 2. Normalized Concentration in the Backfill as a Function of time at Waste-Backfill, $z=0$, and Backfill-Rock Interface, $z=L$.

zero at early time because it takes time for nuclides to diffuse to the interface. At later times the concentration at the interface becomes almost the same as the concentration at the waste surface. This means the concentration in the inner layer has become flat at later time. This is different from the case of the concentration boundary condition problem [2], which showed the concentration at $z=L$ to be greater than that at the $z=0$ for some nuclides due to the interior maximum as mentioned in the introduction.

Figure 3 shows normalized flux profiles at both ends of the inner layer as a function of time. The solid curves represent the fluxes at the waste surface and the dashed ones represent the fluxes at the outer interface. The trend of the flux at the outer interface is the same as the trend of the concentration at that point. The mass flux of ^{234}U at $z=0$ becomes almost equal to that at $z=L$ for $t > 1000$ years, which means the backfill can no longer retard the migration of uranium. But ^{230}Th does not show this behavior, and the flux at $z=L$ is smaller than that at $z=0$. The backfill still provides some degree of retardation effect for ^{230}Th . The flux of ^{226}Ra at $z=L$ is somewhat higher than that at $z=0$ for $t > 10^6$ years. This is because the retardation coefficient of ^{230}Th is higher than that of ^{226}Ra . Therefore the production rate of ^{226}Ra inside the backfill is greater than that in the waste.

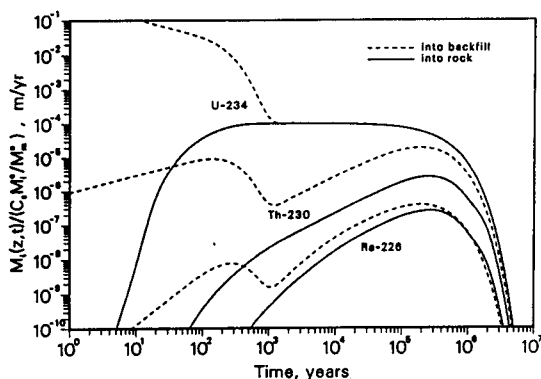


Fig. 3. Normalized Radionuclide mass Fluxes into the Backfill, $z=0$, and into the Rock, $z=L$, as a Function of time.

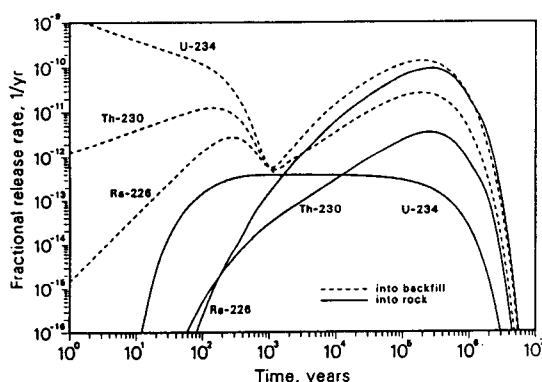


Fig. 4. Fractional Release Rates of Radionuclides Normalized to Their 1000 year Inventories as a Function of time.

Figure 4 shows fractional release rates of the nuclides at both ends of the backfill. The fractional release rates are based on their 1000-year inventories, with the solubility of uranium, $\alpha = 10^{-3} \text{ g/cm}^3$. In this figure the solid curves represent the fractional release rates at the inner interface and the dashed ones represent the fractional release rates at the outer interface. Because the values in this figure are based on 1000-year inventories, the fractional release rates at 1000 years at the interface are identical for all nuclides, by the definition of congruent release. Because of the continued growth of ^{226}Ra and ^{230}Th in the waste

solid, the release rates of these species increase with time. Because of this ingrowth and the 1000-year normalization, the post-1000-year fractional release rates of ^{226}Ra and ^{230}Th are greater than that of ^{234}U . The trends of the fractional release rates in to the outer layer are the same as those into the inner layer except at early times.

4. Conclusions

The general non-recursive solutions for the mass transport of a radioactive decay chain in the finite medium are obtained when nuclides are released congruently with the matrix species. Before this analysis became available, the concentration boundary condition was used to analyze the mass transport of the radionuclide. In the analysis of the concentration boundary condition, however, there were unresolved problems, e. g., the interior maximum in the concentration profile which exceeds the solubility limit, or the negative mass flux at the waste surface. In the problems, e.g., the interior maximum in the

To show numerical illustrations the decay chain $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ in a spent fuel waste is used. This is an important chain in the safety assessment of radioactive waste repositories. However the solutions are not limited by this choice, and can handle a decay chain of an arbitrary number of chain members.

These solutions may be useful and potentially important in performance assessment of radioactive waste repositories.

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