MIGRATION OF RADIONUCLIDES THROUGH SORBING MEDIA ANALYTICAL SOLUTIONS - I

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REPORT OUTLINE

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ANALYTICAL SOLUTIONS - I

Acknowledgments

1. Introduction

2. The Radionuclide Transport Equation
   2.1. Introduction
   2.2. Transport Equation for Radionuclides in Porous Media
       2.2.1. General transport equation in porous media
       2.2.2. The equation of continuity
   2.3. Governing Equations for Radionuclide Migration
       2.3.1. Simplifying assumptions
       2.3.2. Transport with local chemical-sorptive equilibria
       2.3.3. One-dimensional transport with local chemical-sorptive equilibria
       2.3.4. One-dimensional transport without local chemical equilibrium
       2.3.5. Transport along interfaces

3. Nuclide Release Modes
   3.1. Introduction
   3.2. Nuclide Release Modes
       3.2.1. Constant rate of dissolution of waste material
       3.2.2. Nuclide-dependent fractional area of dissolution

3.3 Boundary Conditions and Source Terms

3.4. General Superposition Equation for Band Release (F1)
4. Recursive and General Solutions of the Transport Equation for Nuclide Migration

4.1. Introduction

4.2. Solution of the Transport Equation with a Nuclide Source
   4.2.1. Recursive solution of the transport
   4.2.2. Concentration of the \( i \)th nuclide in a decay chain

4.3. Recursive Solution of the Transport Equation with a Concentration Boundary Condition (P. L. Chambré)
   4.3.1. Integral transform method
   4.3.2. The Green's function method

4.4. Recursive and General Solutions of the Transport Equation without Dispersion (P. L. Chambré)
   4.4.1. Recursive solution of the transport equation without dispersion and some properties of the solution
      A. Integral Transform Method
      B. Classical Method
   4.4.2. The general solution of the transport equations without dispersion

4.5. Local Secular Equilibrium in a Porous Medium

5. Application of General Solutions of the Transport Equation
   5.1. Introduction
5.2. Application of the General Solution of the Transport Equation Without Dispersion—Solutions for Several Release Modes

5.2.1. Solution for preferential release
5.2.2. Solution for exponential release
5.2.3. Solution for step release
5.2.4. Solution for bang release
5.2.5. Solution for impulse release

5.3. Application of the General Solution—Solution of the Transport Equation for a Three-Member Decay Chain with Dispersion, Using the Plane-Source Boundary Condition (H1)

5.4. Application of the General Solution—Solution of the Transport Equation for a Three-Member Decay Chain with Dispersion, Using the Concentration Boundary Condition (F2)

5.5. Migration Behavior of the Decay Chain $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ (H4)

5.5.1. Input data and parameters

5.5.1.1. Release mode
5.5.1.2. Data

5.5.2. Concentration profiles of the Decay Chain $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$

5.5.2.1. Comparison of plane-source and concentration boundary conditions, and effect of dispersion
5.5.2.2. Concentration profiles of nuclides in the case of a pure $^{234}\text{U}$ source in the waste at $t = 0$
5.5.2.3. Concentration profiles of nuclides in the case of transient equilibrium at $^{234}$U daughters in the waste at $t = 0$

5.5.3. Maximum concentration and isopleths of $^{226}$Th

5.5.4. Parametric study

5.5.4.1. The effect of the sorption retardation coefficient on the concentration profiles of $^{226}$Ra

5.5.4.2. The effect of sorption retardation coefficient on the maximum discharge rate of $^{226}$Ra

5.5.4.3. Effect of leach time on the maximum discharge rate of $^{226}$Ra

5.6. Migration Behavior of the 4n+1 Decay Chain

5.6.1. Reduction to a three-member decay chain

5.6.2. Discharge-rate profiles of the $^{237}$Np $\rightarrow$ $^{233}$U $\rightarrow$ $^{229}$Th ($\rightarrow$ $^{225}$Ra) decay chain

6. Nuclide Migration in a Multi-Layered Geologic Medium

6.1. Introduction

6.2. Transport Equation and Conditions at the Layer Boundary

6.3. Analytical Solutions for the Non-Dispersion Case
6.4. Application of the Recursive Solution to a Two-Medium System

6.4.1. General release mode at the repository: $^1\psi_i(t)$

6.4.2. Step release

6.4.3. Step-release solution in a two-medium system for a three-member chain, no dispersion

6.5. Recursive Formula for a Two-Medium System with Dispersion

7. Conclusions

8. Nomenclature

9. References

Appendix A. Computer Code MGRATO3 (UCB-NE-10)

A.1 Program Description

A.1.1. Overall Structure

A.1.2. Input/Output

A.1.2.1. Input

A.1.2.2. Output

A.1.3. Calculation of $E(i,j;k)$

A.1.4. Three-member solution for the preferential-release mode

A.1.5. Program listing of MGRATO3 (UCB-NE-10)

A.1.6. Input data card instruction

A.1.7. Sample of OUTPUT
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1. INTRODUCTION

This report presents analytical solutions for the transport of radionuclides through porous sorbing media. These solutions are developed to serve as analytical standards for benchmark checking of numerical solutions and to use as predictive techniques.

The basic transport equation is derived and is simplified for the case of one-dimensional transport through isotropic porous media, with local chemical equilibria between radionuclides in water solution and sorbed by the solid. Analytical solutions in the present report are limited to this simplified model and are also limited to an infinite plane source of dissolving radionuclides.

Possible modes of release of radionuclides from the waste solid into the ground water are examined. Release-mode equations and boundary conditions are developed for the cases of (a) a constant overall rate of dissolution of the waste matrix and its contained radionuclides, and (b) a constant fractional rate of dissolution of each radionuclide species. Each release mode can be written in terms of a concentration boundary condition for each radionuclide at the point of dissolution, which is applicable if transport by liquid diffusion at the waste location can be neglected, or, more generally, as a volumetric source term to be used directly in the transport equation.

A finite dissolution rate of the waste material results in a "band release", wherein a chromatographic band of the released radionuclides propagates outward through the geologic medium. In Chapter 3 a superposition theorem is developed such that the relatively complicated
equations for band release can be constructed from the simpler solutions for a "step release", which assumes that the waste material dissolves at a finite rate over an infinite period of time.

In Chapter 4, recursive and general solutions of the transport equation, with and without dispersion, are presented. Solutions are presented for individual members in a three-member decay chain and also for any member in a decay chain involving an arbitrary number of members.

In Chapter 5 the recursive and general solutions are applied to the various release modes, including the limiting cases of impulse release and step release, for an infinite geologic medium surrounding the radioactive waste. The solutions are demonstrated for the three-member chain

\[ ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra} \]

which is of practical importance in the analysis of possible long-term environmental releases from high-level waste repositories. Effects of the alternative source boundary conditions are demonstrated. The properties of the maximum concentration of Ra-226, its increase with distance from the repository, and its relative insensitivity to the dissolution rate, are illustrated.

The analytical solutions are also demonstrated for the radionuclide chain

\[ ^{237}\text{Np} \rightarrow ^{233}\text{Pa} \rightarrow ^{233}\text{U} \rightarrow ^{229}\text{Th} \rightarrow ^{225}\text{Ra} \]

which is also important in analyses of long-term environmental effects.

In Chapter 6 the general solutions are applied to multi-layered geologic media and explicitly for a two-medium system consisting of a
medium of finite thickness adjacent to the radioactive waste, sandwiched between infinite half spaces of a second medium. Explicit solutions are developed for a three-member decay chain without dispersion. A recursive formula is developed for the system with dispersion.

In the Appendix a simple computer program is presented, written to demonstrate the migration features of radionuclides in a three-member chain, with and without dispersion.

The analyses presented herein were developed in part under financial support from the University of California and were extended under financial support from the Office of Waste Isolation (ONWI) of the U.S. Department of Energy. Work under ONWI support, performed during the period April 1 through August 31, 1979, is included in this report. The ONWI Project Officer is Dr. H. C. Burkholder.

More detailed demonstration of the use of these equations, as well as analytical solutions for non-equilibrium sorption and multi-dimensional cases, will be the subject of future reports.
2. THE RADIONUCLIDE TRANSPORT EQUATION

2.1 Introduction

The accurate prediction of the probable pattern of contaminant migration is of primary concern in the evaluation of geological disposal of high-level radioactive waste. This chapter is concerned with the formulation of the basic differential equation for the transport of radionuclides in a geologic medium saturated with groundwater. Section 2.2 contains a discussion of the transport equation for porous media. In Section 2.3 a linearized formulation is discussed.

2.2 Transport Equation for Radionuclides in Porous Media

In this section the equations for transport of radionuclide through porous media will be developed with the aid of the averaging concept for a heterogeneous water-solid medium, as proposed by Whitaker (W1) and Slattery (S2).

2.2.1 General transport equation in porous media

The geological medium is considered to be a composite material which consists of fluid and solid phases. The phenomena of nuclide migration through a geological medium are complicated microscopically because the porous medium is not an ordered structure. We can formulate the transport equation governing the nuclide migration in a geological medium according to a statistical approach (W1,S2) which smooths out the properties in complicated composite material by averaging over a small volume domain in this medium.
Let us consider a composite medium as shown in Figure 2.1, in which arbitrary positions are measured by the reference coordinate. The volume of an arbitrary domain D is designated by V. The domain is fixed in time and in space. The center of the domain occupies the point 0', the position of which is designated as X on the reference coordinate. The position of arbitrary point in the domain D is designated as x, the relative position vector from the point 0'.

We can select the domain D of a volume element which is small enough compared to the characteristic length of the composite medium and large enough compared to the microscopic feature of the composite medium, e.g., the inter-particle pore size.\textsuperscript{a1}

Consider a dynamical property, \( \psi \), which is generally described as

\[
\psi = \psi(X, x, t)
\]

The transport equation which governs the time-dependent and spatially-dependent properties in the composite medium system can be obtained by the following three steps:

1. Smooth the property in the domain with the aid of volume averaging:

\[
\hat{\psi} = \frac{1}{V} \psi(X, x, t)
\]

\textsuperscript{a1}/ In this system, there are two kinds of pores. One is a pore into which fluid can penetrate. The other is a closed pore in the solid phase, into which water cannot penetrate. The latter is included as part of the solid phase.
Fig. 2.1 - The averaging volume domain comprising the solid phase and fluid phase.
2. Assign the smoothed value to the property at the center of the domain. The property is thus reduced to:
\[
\hat{\psi} = \hat{\psi}(X,t)
\]  
(2.3)

\(\hat{\psi}\) is to be considered as a point function with respect to the position \(X\).

3. Derive the transport equation for the point function, .

The values of the dynamical property are different for solid and fluid phases, \(s\) and \(f\), in the domain.

\[
\psi(X,x,t) = \begin{cases} 
\psi_f(X,x,t) & , \ x \in x_f \\
\psi_s(X,x,t) & , \ x \in x_s
\end{cases}
\]  
(2.4)

where \(x_\alpha\) denotes the position in the phase \(\alpha\). We can define a function as:

\[
H(x \in x_\alpha) = \begin{cases} 
1 & , \ x \in x_\alpha \\
0 & , \ x \notin x_\alpha
\end{cases}
\]  
(2.5)

From the above definition, the \(\psi\) values in each phase and in the entire domain can be defined as:

\[
\psi_\alpha(X,x,t) \equiv \psi_\alpha H(x \in x_\alpha)
\]  
(2.6a)

\[
\psi_\alpha(X,x,t) \equiv \psi_s H(x \in x_s) + \psi_f H(x \in x_f)
\]  
(2.6b)
The volume average for (2.6) is defined as:

\[ \hat{\psi}_\alpha = \frac{1}{V} \int_V \psi_\alpha \, dx = \frac{1}{V} \int_{V_\alpha} \psi_\alpha \, dx = \varepsilon_\alpha \bar{\psi} \]  

(2.7a)

\[ \hat{\psi} = \frac{1}{V} \int_V \psi \, dx = \hat{\psi}_f + \hat{\psi}_s \]  

(2.7b)

where

\[ \bar{\psi}_\alpha = \frac{1}{V_\alpha} \int_{V_\alpha} \psi_\alpha \, dx \]  

(2.8)

and

\[ \varepsilon_\alpha = \frac{V_\alpha}{V} = \text{volume fraction of the } \alpha \text{ phase.} \]  

(2.9)

The average, \( \bar{\psi} \), is an intrinsic value for the \( \alpha \)-phase. \( V_\alpha \) is the volume of the \( \alpha \)-phase in the domain and is generally a function of time.

Next we will find the averaging rules for the time derivative and the position derivative with respect to \( X \), which usually appear in the transport equation for a homogenous medium. Differentiating (2.7a) with respect to time under constant volume, \( V \), we obtain

\[ \frac{\partial \hat{\psi}_\alpha}{\partial t} = \frac{1}{V} \int_V \frac{\partial \hat{\psi}}{\partial t} H(x \in x_\alpha) \, dx \]  

\[ + \frac{1}{V} \int_V \psi_\alpha \frac{\partial H(x \in x_\alpha)}{\partial t} \, dx \]  

(2.10)
We can use the following equation:

\[
\frac{\partial H(x \in x_\alpha)}{\partial t} = \left( \frac{\partial}{\partial x_b} \right) H(x \in x_\alpha) \cdot \frac{dx_b}{dt} = \delta(x_b - x)w \cdot n_\alpha
\]  

(2.11)

where \(x_b\) denotes the position on the interface between the solid and fluid phases, \(n_\alpha\) is the unit normal vector pointing out of the interface from the \(\alpha\)-phase, \(w\) is the velocity of the interface, and \(\delta(x)\) is the Dirac delta function. The second term of (2.10) is rewritten with the aid of (2.11) as:

\[
\frac{1}{V} \int_V \psi_\alpha \frac{\partial H(x \in x_\alpha)}{\partial t} \, dx = \frac{1}{V} \int_V \psi_\alpha \delta(x_b - x)w \cdot n_\alpha \, dx
\]

\[
= \frac{1}{V} \int_A \psi_\alpha(x_b, t)w \cdot n_\alpha \, dx_b
\]  

(2.12)

where \(A\) is the interfacial area within the domain.

Introducing (2.12) into (2.10), we have:

\[
\frac{\partial \hat{\psi}_\alpha}{\partial t} = \frac{\hat{\partial} \psi_\alpha}{\partial t} + \frac{1}{V} \int_A \psi_\alpha(x_b, t)w \cdot n_\alpha \, dx_b
\]  

(2.13)

Writing (2.13) for each phase and adding, we obtain

\[
\frac{\partial \hat{\psi}}{\partial t} = \frac{\hat{\partial}(\psi_s + \psi_f)}{\partial t} + \frac{1}{V} \int_A [\psi_s(x_b, t) - \psi_f(x_b, t)]w \cdot n_s \, dx_b
\]  

(2.14)

Equations (2.13) and (2.14) are the averaging rules for the time derivative.
We can derive the averaging rule for the position derivative from a procedure similar to the above. The dynamical property $\psi$ can be written as:

$$\psi(X, x, t) = \psi(X + x, t) \quad (2.15)$$

because $\psi$ is a point function with respect to the position vector. $\psi$ is rewritten with use of the function defined by (2.5) as:

$$\psi_\alpha(X, x, t) = \psi_\alpha(X + x, t) \cup [(X + x) \in (X + x)]$$

$$\psi_\alpha(X, x, t) = \psi_f(X + x, t) + \psi_s(X + x, t) \quad (2.16)$$

The value of $\psi_\alpha$ integrand over the fixed domain is now differentiated with respect to the position vector $X$.

$$\nabla_X \int_V \psi_\alpha H \{ (X + x) \in (X + x) \} \text{d}x$$

$$= \int_V \nabla_X \{ \psi_\alpha H \{ (X + x) \in (X + x) \} \} \text{d}x$$

$$= \int_V \nabla_X \{ \psi_\alpha H \{ (x + X) \in (x + X) \} \} \text{d}x$$

$$= \int_V (\nabla_X \psi_\alpha) H \{ (x + X) \in (x + X) \} \text{d}x + \int_V \psi_\alpha \nabla_X H \{ (x + X) \in (x + X) \} \text{d}x$$

$$= \int_V (\nabla_X \psi_\alpha) H \{ (x + X) \in (x + X) \} \text{d}x - \int_V \psi_\alpha \delta(x_b - x)n_\alpha \text{d}x$$

$$= \int_V \nabla_X \psi_\alpha \text{d}x - \int_A \psi_\alpha (x_b, t)n_\alpha \text{d}x_b$$

$$\quad (2.17)$$
Dividing both hand-sides by \( V \), the volume average can be obtained as

\[
\nabla_x \hat{\psi}_\alpha = \nabla_x \psi_\alpha - \frac{1}{V} \int_A \psi_\alpha(x_b,t)n_\alpha \, dx_b
\]

(2.18)

For \( \psi \),

\[
\nabla_x \hat{\psi} = \nabla_x \psi - \frac{1}{V} \int_A \{ \psi_s(x_b,t) - \psi_f(x_b,t) \} n_s \, dx_b
\]

(2.19)

These two equations (2.1) relate the derivative of the averaged value to the average of the derivative of \( \psi \). Henceforth \( \psi_f(x_b,t) \) is simply designed at \( \psi_f(x_b) \).

As a first contribution to the transport equation, the material derivative of the dynamical property is given by:

\[
\frac{d}{dt} \int_V \psi_\alpha \mathbf{H}(x \in \chi_\alpha) \, dx = \frac{d}{dt} \int_{V_\alpha(t)} \psi_\alpha \, dx
\]

\[
= \int_{V_\alpha} \frac{\partial \psi_\alpha}{\partial t} \, dx + \int_{S_\alpha} \psi_\alpha \mathbf{v}_\alpha \cdot n_\alpha \, dS + \int_A \psi_\alpha(x_b) \mathbf{v}(x_b) \cdot n_\alpha \, dx_b
\]

(2.20)

where \( S_\alpha \) is the exit or entrance surface area enclosing the phase \( \alpha \) in the domain. \( \mathbf{v}_\alpha(x_b) \) is the velocity for the \( \alpha \)-phase at the interface. From (2.20) to (2.13), we can rewrite the general transport equation as:

\[
\frac{d}{dt} \int_V \psi_\alpha \mathbf{H}(x \in \chi_\alpha) \, dx = \frac{\partial}{\partial t} \int_V \psi_\alpha \mathbf{H}(x \in \chi_\alpha) \, dx + \int_A \psi_\alpha(x_b) [\mathbf{v}(x_b) - \mathbf{\omega}] \cdot n_\alpha \, dx_b + \int_{S_\alpha} \psi_\alpha \mathbf{v}_\alpha \cdot n_\alpha \, dS
\]

\[
= \text{birth rate of } \psi_\alpha \text{ in the } \alpha \text{-phase}
\]

(2.21)
The left-hand side of this equation is equal to the birth rate of $\psi_\alpha$ in the $\alpha$-phase of the domain. Using the Gauss theorem, this equation can be rewritten as:

$$
\frac{d}{dt} \int_V \psi_\alpha H(x \in x_\alpha) dx + \int_{V_\alpha} \nabla \cdot (\psi_\alpha \mathbf{v}_\alpha) dx - \int_A \psi_\alpha (x_b) \mathbf{w} \cdot \mathbf{n} \alpha dx_b
$$

= birth rate of $\psi_\alpha$ in $\alpha$-phase

(2.22)

For $\psi_\alpha = \psi_f + \psi_s$ within the domain we have:

$$
\frac{d}{dt} \int_V \psi_\alpha dx = \frac{d}{dt} \int_V \psi_f dx + \int_V \nabla \cdot (\psi_s \mathbf{v}_s + \psi_f \mathbf{v}_f) dx - \int_A (\psi_\alpha (x_b) - \psi_f(x_b)) \mathbf{w} \cdot \mathbf{n} s dx_b
$$

= birth rate of $\psi$ in the whole space of the domain

(2.23)

For a mixture of $m$ chemical species, consider $m$ continuous bodies, each of which is visualized by the region it occupies in the physical space. The bodies are allowed to occupy a common portion of the physical space. In this case, the material derivative of the dynamical property for the $k$th chemical species is written by

$$
\frac{d}{dt} \int_V \psi_{k\alpha} H(x \in x_\alpha) dx = \frac{d}{dt} \int_{V_\alpha} \psi_{k\alpha} dx
$$

= \int_{V_\alpha} \frac{\partial \psi_{k\alpha}}{\partial t} dx + \int_{S_\alpha} \psi_{k\alpha} \mathbf{v}_{k\alpha} \cdot \mathbf{n}_\alpha ds + \int_A \psi_{k\alpha} \mathbf{v}_{k\alpha} (x_b) \mathbf{n}_\alpha dx_b
$$

(2.24)
where subscript \( k \) denotes the value for the \( k^{th} \) species. Introduction of (2.13) into (2.24) gives the transport equation for the \( k^{th} \) species as:

\[
\frac{\partial}{\partial t} \int_{V_\alpha} \psi_{k\alpha} \, dx + \int_{V_\alpha} \nabla \cdot (\psi_{k\alpha} \mathbf{v}_{k\alpha}) \, dx - \int_{A} \psi_{k\alpha} \mathbf{w} \cdot \mathbf{n} \, dx_b = \text{birth rate of } k^{th} \text{ species in } \alpha\text{-phase} \equiv R_{k\alpha}
\]

(2.25)

Equations (2.22) and (2.25) can be rewritten with help of (2.18) as:

\[
\frac{\partial}{\partial t} \left( \epsilon \tilde{\psi}_{\alpha} \right) + \nabla \cdot (\tilde{\psi}_{\alpha} \mathbf{v}_{\alpha} \epsilon_{\alpha}) + \frac{1}{V} \int_{A} \psi_{\alpha}(\mathbf{x}_b)[\mathbf{v}_{\alpha}(\mathbf{x}_b) - \mathbf{w}] \cdot \mathbf{n} \, dx_b = \text{birth rate of } \psi_{\alpha} \text{ in the } \alpha\text{-phase}
\]

(2.26)

\[
\frac{\partial}{\partial t} \left( \epsilon \tilde{\psi}_{k\alpha} \right) + \nabla \cdot (\tilde{\psi}_{k\alpha} \mathbf{v}_{k\alpha} \epsilon_{\alpha}) + \frac{1}{V} \int_{A} \psi_{k\alpha}(\mathbf{x}_b)[\mathbf{v}_{k\alpha}(\mathbf{x}_b) - \mathbf{w}] \cdot \mathbf{n} \, dx_b = \text{birth rate of } \psi_{k\alpha} \text{ in the } \alpha\text{-phase}
\]

(2.27)

In the above derivation we have used the relation:

\[
\frac{1}{V} \int_{V} \psi_{\alpha} H(\mathbf{x} \in \mathbf{x}_{\alpha}) \, dx = \frac{1}{V} \int_{V_{\alpha}} \psi_{\alpha} \, dx = \epsilon \tilde{\psi}_{\alpha}
\]

(2.28)

Equation (2.27) can be rewritten by introducing the relative flux \( j_k \) of the property \( \psi \) of the \( k^{th} \) species within the \( \alpha \)-phase, which moves at a velocity \( (\mathbf{v}_{k\alpha} - \mathbf{v}_{\alpha}) \) relative to the \( \alpha \)-phase, i.e.,

\[
j_{k\alpha} = \psi_{k\alpha}(\mathbf{v}_{k\alpha} - \mathbf{v}_{\alpha})
\]

(2.29)
If the solid phase is stationary, \( j_{k\alpha} \) measured at the solid-fluid interface is the net flux across the interface. Substituting (2.29) into (2.27) results in:

\[
\frac{\partial}{\partial t} (e_{\alpha \psi_{k\alpha}}) + \nabla \cdot (e_{\alpha \psi_{k\alpha}} \mathbf{v})
\]

\[
+ \frac{1}{V} \int_{A} [j_{k\alpha}(x_b) + \psi_{k\alpha}(x_b) \{ \mathbf{v}_{\alpha}(x_b) - \mathbf{w} \}] \cdot \mathbf{n}_{\alpha} \, dx_b
\]

\[= -\nabla \cdot (e_{\alpha \mathbf{j}_{k\alpha}})_{mkd} + \text{birth rate of } \psi_{k\alpha} \text{ in a } \alpha\text{-phase} \quad (2.30) \]

(2.26) and (2.30) are the general transport equations for a single body and for multi-bodies of a mixture, respectively.

These equations can also be obtained by integrating the single-phase transport equations over the volume domain and by using the averaging rules (2.13) and (2.18).

2.2.2 The equation of continuity

In the case when \( \psi \) is selected as the density of the \( \alpha \)-phase, the birth rate of \( \psi \) becomes zero, because radioactive decay contributes negligibly to a mass charge. Then, from (2.26),

\[
\frac{\partial}{\partial t} (e_{\alpha \rho_{\alpha}}) + \nabla \cdot (e_{\alpha \rho_{\alpha}} \mathbf{v}) + \frac{1}{V} \int_{A} \rho_{\alpha} (\mathbf{v}_{\alpha} - \mathbf{w}) \cdot \mathbf{n}_{\alpha} \, dx_b = 0 \quad (2.31)
\]

This equation is the mass-conservation equation for the \( \alpha \)-phase. The last term denotes the rate of change of the \( \alpha \)-phase to another phase. The sum of (2.31) with respect to both solid and fluid phases yields
\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\hat{\rho} \hat{\mathbf{v}}) = 0
\]  
(2.32)

where

\[
\hat{\rho} = \varepsilon_s \hat{\rho}_s + \varepsilon_f \hat{\rho}_f, \quad \hat{\mathbf{v}} = \varepsilon_s \hat{\mathbf{v}}_s + \varepsilon_f \hat{\mathbf{v}}_f
\]

and we have used the continuity equation at the interface:

\[
\rho_f [\mathbf{v}_f(x_b) - \mathbf{w}] \cdot n_f \, dx_b + \int_A \rho_s [\mathbf{v}_s(x_b) - \mathbf{w}] \cdot n_s \, dx_b = 0
\]

Next, we will derive the continuity equation of the \( k^{th} \) chemical species in the mixture. In this case, the general transport Equation (2.30) for the mixture should be used. Selecting the molar concentration \( C_{k\alpha} \) as \( \psi_{k\alpha} \), Equation (2.30) can be written as:

\[
\frac{\partial}{\partial t} (\varepsilon_\alpha \tilde{C}_{k\alpha}) + \nabla \cdot (\varepsilon_\alpha C_{k\alpha} \hat{\mathbf{v}}) + \nabla \cdot (\varepsilon_\alpha \tilde{j}_{k\alpha})
\]

\[+ \frac{1}{V} \int_A [j_{k\alpha}(x_b) + C_{k\alpha}(x_b) \{ \mathbf{v}_\alpha(x_b) - \mathbf{w} \}] \cdot n_\alpha \, dx_b = \hat{R}_{k\alpha}
\]  
(2.33)

where

\[
\hat{R}_{k\alpha} = \frac{1}{V} \int_{V_\alpha} R_{k\alpha} \, dx
\]

and \( R_{k\alpha} \) is the birth rate of the \( k^{th} \) species in the \( \alpha \)-phase.

The second term in (2.33) represents convective transport, the third term represents diffusive transport, and the fourth term represents the net flow of the \( k^{th} \) species across the solid-fluid interface. Summing over \( k \), (2.33) is reduced to (2.31), which is the continuity equation for total mass in the \( \alpha \)-phase.
We denote the deviations of $C_{k\alpha}$ and $v_\alpha$ from their averaged values in the domain as $\tilde{C}_{k\alpha}$ and $\tilde{v}_\alpha$, respectively. Then,

$$C_{k\alpha} = \tilde{C}_{k\alpha} + \dot{C}_{k\alpha}, \quad v_\alpha = \tilde{v}_\alpha + \dot{v}_\alpha$$

(2.34)

Each deviation is reduced to zero if averaged:

$$\int_{V_\alpha} \tilde{C}_{k\alpha} \, dx = \int_{V_\alpha} \tilde{v}_\alpha \, dx = 0$$

(2.35)

Using (2.34) and (2.35), the term $\nabla \cdot (\epsilon_{\alpha} \tilde{C}_{k\alpha} \tilde{v}_\alpha)$ in (2.33) becomes:

$$\nabla \cdot (\epsilon_{\alpha} \tilde{C}_{k\alpha} \tilde{v}_\alpha) = \nabla \cdot (\epsilon_{\alpha} \tilde{C}_{k\alpha} \tilde{v}_\alpha) + \nabla \cdot (\epsilon_{\alpha} \tilde{C}_{k\alpha} \tilde{v}_\alpha)$$

(2.36)

Introduction of (2.36) into (2.33) yields:

$$\frac{\partial}{\partial t} (\epsilon_{\alpha} \tilde{C}_{k\alpha}) + \nabla \cdot (\epsilon_{\alpha} \tilde{C}_{k\alpha} \tilde{v}_\alpha) + \nabla \cdot [\epsilon_{\alpha} \tilde{j}_{k\alpha} + \epsilon_{\alpha} \tilde{C}_{k\alpha} \tilde{v}_\alpha]$$

$$+ \frac{1}{V} \int_A [j_{k\alpha}(x_b) + C_{k\alpha}(x_b) (v_\alpha(x_b) - w)] \cdot n_\alpha \, dx_b = \frac{1}{V} \int_{V_\alpha} R_{k\alpha} \, dx$$

(2.37)

Here the mass flux $j_{k\alpha}$ is the net molecular flux for the $k^{th}$ chemical species relative to the motion of the $\alpha$-phase. In this work, the general constitutive relations governing $j_{k\alpha}$ will not be treated. Instead, we assume Fick's law of diffusion:

$$j_{k\alpha} = -D_{k\alpha} \nabla_x C_{k\alpha}$$

(2.38)

With the help of (2.38) and (2.18) Equation (2.37) is reduced to:
\[
\frac{\partial}{\partial t} \left( \varepsilon_{\alpha} \tilde{c}_{k\alpha} \right) + \nabla \cdot \left( \varepsilon_{\alpha} \tilde{c}_{k\alpha} \tilde{v}_{\alpha} \right) + \nabla \cdot \left( \varepsilon_{\alpha} \tilde{v} \tilde{c}_{k\alpha} \right) \\
- \nabla \cdot \left[ D_{k\alpha} \nabla \left( \varepsilon_{\alpha} \tilde{c}_{k\alpha} \right) \right] - \nabla \cdot \frac{1}{V} \int_{A} D_{k\alpha} \rho_{\alpha} n_{\alpha} d\mathbf{x}_b \\
+ \frac{1}{V} \int_{A} \left[ j_{k\alpha} (\mathbf{x}_b) + \rho_{k\alpha} (\mathbf{x}_b) \left( \mathbf{v}_{\alpha} (\mathbf{x}_b) - \mathbf{w} \right) \right] \cdot n_{\alpha} d\mathbf{x}_b = \varepsilon_{R_{k\alpha}}
\] (2.39)

where the spatial change of $D_{k\alpha}$ in the averaging domain was neglected.

The fifth term in the left hand side of (2.39) means a kind of "tortuosity" effect (S2,G1) which yields the reduction of diffusion flow.

We are primarily concerned here with the migration of radionuclides, which may exist in the ground water in various chemical forms. Let us consider a decay chain such that the $i^{th}$ nuclide species within a given phase exists in $m$ different chemical forms, each form $k$ containing $S_i^k$ atoms of nuclide $i$ per molecule.

The birth rate of the $i^{th}$ nuclide takes the form

\[
\sum_{k=1}^{m} S_i^k \lambda_{i-1} \sum_{k=1}^{m} \tilde{S}_i^{k-1} \rho_{k\alpha} - \lambda_{i} \sum_{k=1}^{m} S_i^k \tilde{c}_{k\alpha} + \tilde{f}_{i\alpha}(\mathbf{x},t)
\] (2.40)

where $\tilde{f}_{i\alpha}(\mathbf{x},t)$ is the source term of the $i^{th}$ nuclide in the $\alpha$-phase at the position $\mathbf{x}$ and at time $t$. If we multiply (2.39) by $S_i^k$ and add the resultant equation with respect to $k$, it follows that
\[
\frac{\partial}{\partial t} \left( \epsilon_{\alpha} \bar{N}_{i\alpha} \right) + \nabla \cdot \left( \epsilon_{\alpha} \nabla \bar{N}_{i\alpha} \right) \\
+ \nabla \left\{ \epsilon_{\alpha} \bar{v}_{i\alpha} - \sum_{k=1}^{m} S_{i}^{k} D_{k\alpha} \nabla \left( \epsilon_{\alpha} \bar{C}_{k\alpha} \right) \right\} \\
- \nabla \left\{ \frac{1}{V} \int_{A} \sum_{k=1}^{m} S_{i}^{k} D_{k\alpha} \epsilon_{\alpha} \bar{N}_{i\alpha} \alpha d\mathbf{x}_{b} \right\} \\
+ \frac{1}{V} \int_{A} \left[ \sum_{k=1}^{m} S_{i}^{k} \bar{N}_{i\alpha} \left( \mathbf{x}_{b} \right) + \bar{N}_{i\alpha} \left( \mathbf{x}_{b} \right) \left( \mathbf{v}_{\alpha} \left( \mathbf{x}_{b} \right) - \mathbf{w} \right) \cdot \mathbf{n} \right] \alpha d\mathbf{x}_{b} \\
= \left( \lambda_{i-1} \bar{N}_{i-1,\alpha} - \lambda_{i} \bar{N}_{i,\alpha} \right) \epsilon_{\alpha} + \epsilon_{\alpha} \bar{f}_{i\alpha} \left( \mathbf{x}, t \right) \tag{2.41}
\]

where \( \bar{N}_{i\alpha} \) is the atomic concentration of the \( i \)th nuclide,

\[
\bar{N}_{i\alpha} = \sum_{k=1}^{m} S_{i}^{k} \bar{C}_{k\alpha}
\]

and \( \lambda_{i} \) is the decay constant for the \( i \)th nuclide. This equation is the mass balance for the \( i \)th nuclide and is a general transport equation governing the migration of a nuclide through a geological medium, which will be further developed in the following section.

2.3 Governing Equations for Radionuclide Migration

In this section we simplify the transport equation (2.41), and we develop the governing equation for the nuclide migration process which will be the subject of analyses described in the later chapters.

Because of the complexity of the geological media, we postulate that the radionuclides are transported by ground water moving in a deterministic flow path with a flow velocity given \textit{a priori}. In this case the transport equations for chemical species and radionuclides are given by (2.39) and (2.41).
2.3.1 Simplifying assumptions

We postulate the following assumptions:

1. The fluctuation terms for the convective transport in the water phase, \( \varepsilon_f \dot{\nabla}_f \dot{C}_{kf} \) and \( \varepsilon_f \dot{\nabla}_f \dot{N}_{if} \), are described as a stochastic random process. This assumption is formulated as

\[
\varepsilon_f \dot{\nabla}_f \dot{C}_{kf} = -D_{kf} \nabla_x (\varepsilon_j \dot{C}_{kj}) \quad \varepsilon_f \dot{\nabla}_f \dot{N}_{if} = -D_{df} \nabla_x (\varepsilon_f \dot{N}_{if}) \quad (2.42)
\]

where \( D_{df} \) is the dispersion tensor.

The convective term within the solid phase is assumed to play no role in the transport of radionuclides.

2. The molecular diffusion coefficients in the water phase are assumed to have a value \( D_{mf} \) for each chemical species. This assumption leads to

\[
\frac{1}{V} \nabla_x \int_{V_f} \sum D_{kf} C_{kf} s_{ik} \, dx = \nabla_x (\varepsilon_f D_{mf} \tilde{N}_{if}) \quad (2.43)
\]

This assumption is reasonable, because the molecular diffusion coefficients of several chemical species are of the same order and because transport by molecular diffusion process is less important than transport by dispersive diffusion, as treated in assumption 1.

The molecular diffusion in the solid phase is also assumed to be independent of chemical species, so that

\[
D_{k,s} = D_s \quad (2.44)
\]
3. The tortuosity term is neglected. Because this term usually reduces the flow due to the molecular diffusion, this assumption is conservative for nuclide migration. Scheidegger (S1) has proposed a dispersion tensor in which both convective fluctuation and tortuosity terms are included.

4. The motion of the interface is negligible, and the mass-averaged velocities of the two phases across the interface are assumed to be zero.

\[ w = v_f(x_b) = v_s(x_b) = 0 \]  \hspace{1cm} (2.45)

This assumption is reasonable when there is no phase change between the water and the solid phase.

5. The bulk motion of the solid phase is neglected.

\[ v_s = 0 \]  \hspace{1cm} (2.46)

The transport equations for chemical species and for radionuclides are then given by:

\[ \frac{\partial}{\partial t} (\varepsilon \tilde{C}_f) + \nabla \cdot (\varepsilon \tilde{C}_f \tilde{v}_f) \]

\[ - \nabla \cdot [D_f \nabla (\varepsilon \tilde{C}_f)] + \frac{1}{V} \int_A j_k(x_b) \cdot n_f dx = \varepsilon \tilde{R}_f \]  \hspace{1cm} (2.47a)

\[ \frac{\partial}{\partial t} (\varepsilon \tilde{C}_s) - \nabla \cdot [D_s \nabla (\varepsilon \tilde{C}_s)] - \frac{1}{V} \int_A j_k(x_b) \cdot n_f dx = \varepsilon \tilde{R}_f \]  \hspace{1cm} (2.47b)
\[
\frac{\partial}{\partial t} (\epsilon_f \bar{N}_{if}) + \nabla \cdot (\epsilon_f \bar{v} \bar{N}_{if})
- \nabla \cdot [D_f \nabla (\epsilon_f \bar{N}_{if})] + \frac{1}{V} \int_A \sum_{k} S_{i k}^k(x_b) \cdot n_f dx
= \epsilon_f [\lambda_{i-1} \bar{N}_{i-1,f} - \lambda_{i-1} \bar{N}_{i,f}] + \epsilon_f \bar{f}_{if}(x,t) \tag{2.48a}
\]

\[
\frac{\partial}{\partial t} (\epsilon_s \bar{N}_{is}) - \nabla \cdot [D_s \nabla (\epsilon_s \bar{N}_{is})] + \frac{1}{V} \int_A \sum_{k} S_{1 i}^k(x_b) \cdot n_f dx_b
= \epsilon_s (\lambda_{i-1} \bar{N}_{i-1,s} - \lambda_{i-1} \bar{N}_{i,s}) + \epsilon_s \bar{f}_{is}(x,t) \tag{2.48b}
\]

where

\[D_f = D_{df} + D_{mf}^l\]

and \(l\) is the unit tensor.

2.3.2 Transport with local chemical-sorptive equilibria

A geologic medium suitable for radioactive waste isolation must have a water velocity small enough to isolate the nuclide from the biosphere. With such slow transport, one might expect that local thermochemical equilibrium will exist within each phase and between water and solid phases. To examine the migration features under the limiting conditions that chemical and sorption processes are locally equilibrated, we further assume that

6. At each position \(x\) the sorption equilibrium is described as a linear relation:

\[
\bar{C}_{ks} = k_D k \bar{C}_{kf} \tag{2.49}
\]

where \(k_{D,k}\) is a distribution coefficient for the \(k\)th chemical species.
7. Different chemical forms of a given chemical element are in equilibrium within a given phase \( \alpha \), so that

\[
\tilde{C}_{t, \alpha} = \tilde{C}_{t, \alpha} \quad (2.50)
\]

where \( \tilde{C}_{t, \alpha} \) is the concentration of the \( t \)th chemical species involving the nuclide \( i \) and \( k_{i, \alpha} \) denotes an equilibrium constant.

From (2.49) and (2.50), the overall sorption equilibrium is expressed in terms of nuclide concentrations \( \tilde{N}_{i, f} \) and \( \tilde{N}_{i, s} \) as:

\[
\tilde{N}_{i, s} = \left[ \frac{\sum_k (k_{i, f}^k / k_{i, f})}{\sum_k (k_{i, f}^k / k_{i, f})} \right] \tilde{N}_{i, f}
\]

\[
\equiv K_{D, i} \tilde{N}_{i, f} \quad (2.51)
\]

where \( K_{D, i} \) is the overall distribution coefficient for the \( i \)th nuclide, i.e.,

\[
K_{D, i} = \frac{\text{concentration of nuclide } i \text{ in the solid}}{\text{concentration of nuclide } i \text{ in the liquid}}
\]

The concentrations \( \tilde{N}_i \) are expressed in consistent units (e.g., moles/liter), so that \( K_{D, i} \) is dimensionless.

Summing up (2.48) with respect to phase and using (2.51) the transport equation governing the \( i \)th nuclide migration is given by the single equation:
\[
\frac{\partial}{\partial t} (\varepsilon_f K_i \tilde{N}_{i+1}^{f, i}) + \nabla \cdot \left[ (\varepsilon_f \nabla \varepsilon_f - D_f \varepsilon_f \nabla \varepsilon_f - D_s \nabla (K_i \varepsilon_s)) \tilde{N}_{i+1}^{f, i} \right]
\]
\[
- \nabla \cdot \left[ (D_f \varepsilon_f + D_s K_i \varepsilon_s) \nabla \tilde{N}_{i+1}^{f, i} \right]
\]
\[
= \varepsilon_f K_i \lambda \tilde{N}_i - \varepsilon_f \tilde{N}_i^{f, i} + \varepsilon_s \tilde{N}_i^{s, i} (x, t)
\]  
(2.52)

where

\[
K_i = 1 + \frac{c_{s}}{\varepsilon_f K_i}
\]  
(2.53)

2.3.3 One-dimensional transport with local chemical-sorptive equilibria

For simplicity, we also assume:

8. The deterministic water pathway is one-dimensional, and the net loss of a nuclide lost from the pathway due to transverse diffusion can be neglected. In fact, in a true one-dimensional flow field, with an infinite plane source of dissolving nuclides, there are no concentration gradients normal to the flow direction to cause transverse diffusion. The geologic medium is assumed to be isotropic.

Equation (2.52) is simplified with these assumptions as:

\[
\frac{\partial}{\partial t} (\varepsilon_f K_i \tilde{N}_{i+1}^{f, i}) + \frac{\partial}{\partial z} (\varepsilon_f \nabla A_{i+1}^{f, i}) - \frac{\partial}{\partial z} (D_f \varepsilon_f B_i \frac{\partial}{\partial z} \tilde{N}_{i+1}^{f, i})
\]
\[
= \varepsilon_f K_i \lambda \tilde{N}_i - \varepsilon_f \tilde{N}_i^{f, i} + \varepsilon_s \tilde{N}_i^{s, i}
\]  
(2.54)

where
\[ A_i = 1 - \frac{D_f (\partial \varepsilon_f / \partial z) + D_s (\partial K_{D_i} \varepsilon_s / \partial z)}{\varepsilon_f \varepsilon_f} \]
\[ B_i = 1 + \frac{D_s K_{D_i} \varepsilon_s}{D_f \varepsilon_f} \]

(2.55)

and \( z \) is the distance in the direction of water flow. \( D_f \) is the dispersion coefficient in the fluid phase, and \( v_f \) is the velocity of the water phase in the \( z \)-direction. If diffusion within the solid phase plays no role in the transport of the nuclide, then \( B_i \) becomes unity.

When \( \varepsilon_f, K_{D_i}, K_i \) and \( v_f \) are constant in space and time the above equation is further reduced to

\[ K_i \frac{\partial \tilde{N}_{if}}{\partial t} + v_f \frac{\partial \tilde{N}_{if}}{\partial z} - D_i \frac{\partial^2 \tilde{N}_{if}}{\partial z^2} = K_{i-1} \tilde{N}_{if} - K_i \tilde{N}_{i-1,f} + \tilde{r}_{if} + \tilde{r}_{is} \varepsilon_s \varepsilon_f \]

(2.56)

where \( D_i = D_f B_i \).

Equations (2.54) and (2.56) are the fundamental equations to be solved in the succeeding chapters of this report.

2.3.4 One-dimensional transport without local chemical equilibrium

For future studies it is helpful to formulate the governing equations of nuclide migration in a more general form, allowing for departure from local chemical equilibrium with regard to sorption and chemical reaction processes.
First, we consider the problem of evaluating the mass flux across the fluid-solid interface \( j_k(x_b) \). Mass transfer between two phases arises from sorption processes, and information concerning the sorption mechanism is necessary for a strict formulation of this flux across the interface. Sorption occurs via three steps: diffusion within the water phase, diffusion through fine pores in the solid phase, and absorption or ion-exchange. We can postulate some limiting cases, e.g., a process controlled by diffusion or a process controlled by reaction rate.

In the case of diffusion controlling, we can formulate \( j_k(x_b) \) by the following equation:

\[
 j_k(x_b) = k_{M,k} \left( \tilde{C}_{kf} - \frac{1}{K_{D,k}} \tilde{C}_{ks} \right) n_f \tag{2.57}
\]

where \( k_{M,k} \) is the overall mass-transfer coefficient for the \( k \)th species, which includes the effects of the diffusional resistances in both phases. This equation is not strictly correct but holds approximately if the sorption process is in a near-equilibrium state.

When mass transfer is controlled by reaction rates, the mass flux across the interface is obtained from an assumed first-order reaction rate with respect to the \( k \)th species.

\[
 j_k(x_b) = k_{A,k} \left[ \tilde{C}_{kf} \tilde{C}_{vs} - \frac{1}{K_A} \tilde{C}_{mf} \tilde{C}_{ks} \right] \frac{V_s}{A} n_f \tag{2.58}
\]

where \( k_{A,k} \) is the reaction-rate constant, and \( K_A \) is the reaction equilibrium constant. In the above equation, \( \tilde{C}_{vs} \) and \( \tilde{C}_{mf} \) represent characteristic concentrations, as defined below. For an ion-exchange reaction process,
\[ \tilde{C}_{vs} = \text{the concentration of the ion-exchangeable species, except for the } k^{th} \text{ species, in the solid phase} \]

\[ \tilde{C}_{mf} = \text{the concentration of the ion-exchangeable species, except for the } k^{th} \text{ species, in the liquid phase} \]

(2.59)

For an adsorption process, we can select

\[ \tilde{C}_{vs} = \text{the vacancy site concentration in the solid phase} \]

\[ \tilde{C}_{mf} = 1 \]

(2.60)

There may exist competing sorptive species in each phase, the concentrations of which may be greater than that of the radionuclide. It is, therefore, reasonable to assume that the \( \tilde{C}_{vs} \) and the \( \tilde{C}_{mf} \) are constant, independent of the concentration of the \( k^{th} \) species. In this case, (2.58) reduces to (2.57).

The integral terms in (2.47) and (2.48) are rewritten for the mass-transfer rate of (2.57) as

\[ \frac{1}{V} \int_A j_k(x_b) \cdot n_f^k dx_b = k_M, k a(\tilde{C}_{mf} - \frac{1}{K_{D,k}} \tilde{C}_{ks}) \]

(2.61)

\[ \frac{1}{V} \sum_k \int_A j_b(x_b) \cdot n_f^k S_i dx_b = \sum_k k_M, k a(\tilde{C}_{mf} - \frac{1}{K_{D,k}} \tilde{C}_{k,s}) S_i \]

(2.62)

where \( a \) is the interfacial area per unit volume of the geologic medium.

In general, the values of \( k_M, k \) and \( K_{D,k} \) are dependent upon the chemical form. It is possible that a given nuclide may exist in more than one
chemical form, even within a given phase. In such cases (2.62) cannot be evaluated in terms of the concentration of nuclide i alone. The prediction of the radionuclide migration then requires the simultaneous solution of the transport equations for chemical species. If the radionuclide is in a single chemical form within a given phase, (2.62) can be simplified as

$$\frac{1}{V} \sum_{k} j_k(x_b) \cdot n_f \int_{V}^{k} \frac{k}{S_b} dx_b = k_{M,i} a (\tilde{N}_{if} - \frac{1}{K_{D,i}} \tilde{N}_{is})$$  \hspace{1cm} (2.63)

In this case, the simultaneous equations for transport of the nuclide (2.48) can be solved directly with the help of (2.63).

Next we consider the possibility that a radionuclide may exist in more than one chemical form within a phase. As explained previously, the sorbing process is strongly influenced by the chemical form of the nuclide. If a certain chemical species is difficult to sorb compared with the other chemical species, the chemical species which sorbs less easily moves more rapidly through the geological medium. Some experimental results suggest the importance of different chemical forms upon the migration features for Pu and Am nuclides (F3). This problem is in principle evaluated by solving the simultaneous transport equations for the different chemical forms of a nuclide with appropriate chemical-reaction rates and mass-transfer rates as discussed previously.

Another problem concerned with chemical reaction is the mineralization reaction, i.e., the nuclide may react irreversibly with some component in the soil. The assumption of reversible sorption, i.e.,
ignoring irreversible mineralization, may always be conservative, but the effect of mineralization upon transport requires elucidation (B5).

This mineralization process can also be considered as the problem concerned with different chemical forms of a radionuclide in the solid phase. We consider two chemical forms, one is desorbable and the other is impossible to desorb. In this case, the transport equations (2.47) for the solid phase can be described as:

\[
\frac{\partial}{\partial t} \left( \varepsilon_s \bar{C}_{1s} \right) - \nabla \cdot \left[ D_s \nabla \left( \varepsilon_s \bar{C}_{1s} \right) \right] - \frac{1}{V} \int_A j_{1i}(x_b) \cdot n_f \, dx_b = \varepsilon_s \bar{R}_{1s} \\
\frac{\partial}{\partial t} \left( \varepsilon_s \bar{C}_{2s} \right) - \nabla \cdot \left[ D_s \nabla \left( \varepsilon_s \bar{C}_{2s} \right) \right] = \varepsilon_s \bar{R}_{2s}
\]  
(2.64)

where \( \bar{C}_{1s} \) and \( \bar{C}_{2s} \) are the solid-phase concentrations of the chemical forms of the desorbable nuclide, \( i_1 \), and of the nuclide impossible to desorb, \( i_2 \). In terms of nuclide concentrations, the above equations can be rewritten as:

\[
\frac{\partial}{\partial t} \left( \varepsilon_s \bar{N}_{1s} \right) - \nabla \cdot \left[ D_s \nabla \left( \varepsilon_s \bar{N}_{1s} \right) \right] - \frac{1}{V} \int_A S^{1i}_{i_1}(x_b) \cdot n_f \, dx_b = \varepsilon_s \bar{R}_{1s} \\
\frac{\partial}{\partial t} \left( \varepsilon_s \bar{N}_{2s} \right) - \nabla \cdot \left[ D_s \nabla \left( \varepsilon_s \bar{N}_{2s} \right) \right] = \varepsilon_s \bar{R}_{2s}
\]  
(2.65)

where

\[
S^{1i}_{i_1i_1s} = S^{i-1}_{i_1} \lambda_i^{i-1} \tilde{N}_{i-1s} - \lambda_i^{i} \tilde{N}_{i1s} - k_i^{i} \tilde{N}_{i1s} \\
S^{2i}_{i_2i_2s} = S^{i-1}_{i_2} \lambda_i^{i-1} \tilde{N}_{i-1s} - \lambda_i^{i} \tilde{N}_{i2s} - k_i^{i} \tilde{N}_{i2s}
\]  
(2.66)
The last term of the above equation, $k_{i_1} N_{i_1}$, denotes the rate of mineralization, where we have assumed that the rate can be expressed by a first-order chemical reaction. $S_{i_1}^{i-1}$ is the fraction of the $(i-1)^{th}$ nuclide that is transformed by decay to the desorbable chemical species of the $i^{th}$ nuclide. In this case, (2.65) and (2.48a) are the fundamental equations to be solved, with the interfacial transfer rate given by:

$$\frac{1}{V} \int \sum_{k} i_k(x) \cdot n_f S_{i_1}^{i} d_{i_1} = k_{M,i} a \left( N_{i_1} - \frac{1}{k_{d,i_1}} N_{i_1} \right)$$

Solution of these non-equilibria forms of the transport equation will be a subject of future studies.

2.3.5 Transport along interfaces

The fundamental premise in developing the transport equations in this chapter has been that the geological medium is composed only of solid and liquid phases. Transport of radionuclides along the interface between two phases has been neglected, although this might become important for strongly sorbing species and with very low water velocities. The treatment of interfacial transport is left for future studies.
3. NUCLIDE RELEASE MODES

3.1 Introduction

The analyses herein provide for various possible modes by which radionuclides can be released at the repository (H5). These release modes affect the boundary conditions at the repository, and they thereby affect the migration features of the radionuclides through the geologic media. In Sections 3.2 mathematical models are developed for the various release modes assumed for this study. In Section 3.3 the boundary conditions and source terms for transport conditions and source terms for transport equation are discussed. In Section 3.4 we present a general superposition relation for constructing the "band-release" solutions.

3.2 Nuclide Release Modes

3.2.1 Constant rate of dissolution of waste material

Consider a radioactive waste consisting of radionuclides dispersed through a solid matrix. It is assumed that:

(a) a constant amount of the total waste material dissolves into ground water per unit time,
(b) all of the waste material begins to dissolve at t=0.

Dissolution is complete within a leach time T, and
(c) all radionuclides contained within the waste material go into solution when the waste matrix dissolves.

Assumption (b) results in a "band-release" of radionuclides into the ground water.
For an initial amount \( M_T^0 \) of total waste material, the rate of dissolution \( M_T \) of the total material is

\[
\dot{M}_T = \frac{M_T^0}{T} \quad (0 \leq t \leq T) \tag{3.1}
\]

The rate of dissolution \( \dot{M}_i(t) \) of radionuclide \( i \) in the waste material is then

\[
\dot{M}_i(t) = n_i(t) \dot{M}_T \tag{3.2}
\]

where \( n_i(t) \) is the amount of nuclide \( i \) per unit amount of waste material.

If the radionuclides dissolve uniformly into the water flowing at a volumetric flow rate \( Q \) past the waste material, and if one neglects the diffusional transport of the dissolved radionuclides in the vicinity of the waste material, the concentration \( N_i(t) \) of a dissolved radionuclide at this location \( (z = 0) \) is given by:

\[
N_i(t) = \frac{\dot{M}_i(t)}{Q} \quad , \quad (0 \leq t \leq T) \tag{3.3}
\]

or using (3.1) and (3.2):

\[
N_i(t) = \frac{n_i(t)M_T^0}{QT} \quad , \quad (0 \leq t \leq T) \tag{3.4}
\]

The waste concentration \( n_1(t) \) of a mother nuclide \( (\lambda_0 = 0) \) in a decay chain is related to its initial waste concentration \( n_1^0 \) by:

\[
n_1(t) = n_1^0 e^{-\lambda_1 t} \tag{3.5}
\]
For the daughter nuclide,

\[ n_2(t) = \frac{n_1^0 \lambda_1}{\lambda_2 - \lambda_1} \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) + n_2^0 e^{-\lambda_2 t}, \] \hspace{1cm} (3.6)

and so on, which continues in the form of the Bateman equation for radioactive decay (B1). Therefore, the time-dependent concentrations \( N_i(t) \) in the water phase at the point of dissolution \( z = 0 \) is given by the Bateman equation:

\[
N_i(t) = \begin{cases} 
0 & t < 0 \\
B_i(t) & 0 < t < T \\
0 & t > T 
\end{cases} \hspace{1cm} (3.7)
\]

The above equation can be rewritten as:

\[
N_i(t) = B_i(t) [h(t) - h(t-T)] \hspace{1cm} (3.8)
\]

where \( h(\xi) \) is the Heaviside step function:

\[
h(\xi) = \begin{cases} 
0 & , \ \xi < 0 \\
1 & , \ \xi > 0 
\end{cases}
\]

\( B_i(t) \) can be obtained from the solution of the differential equation:

\[
\frac{dB_i(t)}{dt} = \lambda_{i-1} B_{i-1} - \lambda_i B_i, \quad (i=1,2,\ldots) \hspace{1cm} (3.9)
\]

with the initial condition

\[
B_i(0) = n_i^0 \hspace{1cm} (3.10)
\]

\( N_i^0 \) is the concentration in the water phase at the instant dissolution begins \( (t = 0) \), measured at the dissolution. The solution of Equation (3.9) is the Bateman equation:
\[ B_i(t) = \sum_{m=1}^{i} \left( \frac{N_i^O}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \sum_{n=m}^{i} \frac{e^{-\lambda_n t}}{\prod_{\ell=m, \ell \neq n} (\lambda_\ell - \lambda_n)} \] 

\[ \equiv \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} \]  

(3.11)

where

\[ B_{ij} = \sum_{m=1}^{j} \frac{N_m^O}{\lambda_m} \left( \frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \left/ \prod_{\ell=m, \ell \neq j} (\lambda_\ell - \lambda_j) \right. \]  

(3.12)

the product term in the denominator of Equation (3.12) is defined as unity when \( m = j = i \). The \( B_i(t) \) values for \( i = 1, 2 \) and 3 are written as:

\[ B_1(t) = N_1^O e^{-\lambda_1 t} \]  

(3.13)

\[ B_2(t) = N_2^O e^{-\lambda_2 t} + \frac{N_2^O \lambda_1}{\lambda_2 - \lambda_1} \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) \]  

(3.14)

\[ B_3(t) = N_3^O e^{-\lambda_3 t} + \frac{N_3^O \lambda_2}{\lambda_3 - \lambda_2} \left( e^{-\lambda_2 t} - e^{-\lambda_3 t} \right) \]  

\[ + \lambda_1 \lambda_2 \lambda_3 \left[ \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right] \]  

(3.15)

When the band-release time \( T \) becomes very small and approaches zero, the release mode becomes an impulse release, such that

\[ N_i(t) = N_i^O \delta(t) \]  

(3.16)

where \( \delta(t) \) is the Dirac delta function.
When the band-release time $T$ approaches infinity, the release mode becomes a step release, such that

$$N_i(t) = B_i(t)h(t)$$  \hspace{1cm} (3.17)

On physical grounds, waste material may reasonably be expected to dissolve at an infinitesimal rate in the case of a step release, because for a step release dissolution is presumed to continue for an infinite time, thereby resulting in only differential concentrations of the dissolved radionuclides. However, it is useful to construct step-release solutions which satisfy the boundary condition of finite concentration in Equation (3.17). If these finite concentrations are those determined from the band-release dissolution model of Equation (3.4) with finite $T$, then the resulting relatively simple step-release solutions can be used in the superposition equation (Eq. (3.60)) developed in Section 3.4 to construct the more complicated band-release solutions.

### 3.2.2 Nuclide-Dependent fractional area of dissolution

Here we assume that the waste material and its contained radio-
uclides begin to dissolve into the water at $t = 0$, and we assume that each radionuclide leaches from the waste at a fractional rate $k_i$, which is a constant for a given radionuclide. The amount $M_i(t)$ of radionuclide $i$ within the repository at time $t$ is given by

$$\frac{dM_i}{dt} = -k_i M_i - \lambda_i M_i + \lambda_{i-1} M_{i-1}$$  \hspace{1cm} (3.18)

with the initial condition
\[ M_i(0) = M^O_i \] (3.19)

The solution of (3.18) and (3.19) is obtained as:

\[ M_i(t) = \sum_{m=1}^{i} \frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r M^O_m \sum_{n=m}^{i} \frac{e^{-\Lambda_n t}}{\prod_{\xi=m}^{n} (\Lambda_\xi - \Lambda_n)_{\xi \neq n}} \] (3.20)

where

\[ \Lambda_i = \lambda_i + k_i \] (3.21)

Assuming, as in Section 3.2.1, that the radionuclides dissolve uniformly into the water flowing at a volumetric rate \( Q \) past the waste material, and neglecting the diffusional transport of the dissolved radionuclides in the vicinity of the waste material, the concentration \( N_i(t) \) in the water phase at \( z = 0 \) is given by:

\[ N_i(t) = \sum_{m=1}^{i} \left( \frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \frac{k_i}{k_m} N^O_m \sum_{n=m}^{i} \frac{e^{-\Lambda_n t}}{\prod_{\xi=m}^{n} (\Lambda_\xi - \Lambda_n)_{\xi \neq n}} \] (3.22)

or

\[ N_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\Lambda_j t}, \quad t > 0 \]

where

\[ B_{ij} = \sum_{m=1}^{j} \left( \frac{k_i}{k_m} N^O_m \right) \prod_{r=m}^{i} \lambda_r \prod_{\xi=m}^{j} (\Lambda_\xi - \Lambda_j)_{\xi \neq j} \] (3.23a)

\[ N^O_m = \frac{k M^O_m}{Q} \] (3.23b)
If the effect of dispersion at \( z = 0 \) is neglected, Equation (3.29) reduces to the simple equation.

\[
N(0^+, t) = N_i(t)
\]  \hspace{1cm} (3.30)

This concentration boundary condition has been used in other published analyses of radionuclide migration (B3, 4, 5, L1).

A more fundamental approach, necessary when diffusional transport both upstream and downstream of the repository is to be considered, is to use the dissolution rate as a source term in the transport equation. The rate of dissolution \( \dot{M}_i(0,t) \) is given from the previous solutions by:

\[
\dot{M}_i(t) = N_i(t)Q
\]  \hspace{1cm} (3.31)

The equivalent distributed volumetric source term \( f_i(z, t) \) to be used in the transport equation is

\[
f_i(z, t) = N_i(t) \frac{Q}{S} \delta(z)
\]  \hspace{1cm} (3.32)

where \( S \) is the effective cross-sectional area for water flow associated with the dissolving waste, or

\[
f_i(z, t) = N_i(t) \delta(z)
\]  \hspace{1cm} (3.33)

where \( v \) is the water velocity at the point of dissolution, as illustrated in Figure 3.2.
section we apply these release modes in the development of a generalized source term at the point of dissolution.

3.3 Boundary Conditions and Source Terms

The concentrations $N_1(t)$ obtained in Section 3.2 are the real concentrations $N_1(0, t)$ at the boundary only if transport by diffusion can be neglected at $z = 0$. However, these concentrations $N_1(t)$ can also yield a nuclide source term to be used directly in radionuclide transport equation, a useful approach if diffusional transport at the source is to be considered and/or if the transport equation is to be solved for spatially distributed sources.

The physical picture for the source condition is illustrated in Figure 3.1, in which the dispersion coefficient $D_1$ is assumed to be zero in the up-stream zone and non-zero in the down-stream zone.

![Figure 3.1](https://via.placeholder.com/150)

**Figure 3.1.** Boundary condition at the repository $(z = 0)$, diffusional transport in flow direction only.

In this case, the nuclide balance at the repository for one-dimensional transport, with flow at velocity $v$ in the $z$ direction, yields

$$-D_1 \left[ \frac{\partial N_1(z,t)}{\partial z} \right]_{z=0^+} = v[N_1(t) - N_1(0^+, t)]$$

(3.29)
$N_m^0$ is the concentration of nuclide $m$ in the water phase at $z = 0$, $t = 0$. For three members,

$$N_1(t) = N_1^0 e^{-\lambda_1 t}$$  \hspace{1cm} (3.24)

$$N_2(t) = N_2^0 e^{-\lambda_2 t} + \frac{k_2}{k_1} N_1^0 \frac{\lambda_1}{\lambda_2 - \lambda_1} \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right)$$  \hspace{1cm} (3.25)

$$N_3(t) = N_3^0 e^{-\lambda_3 t} + \frac{k_3}{k_2} N_2^0 \frac{\lambda_2}{\lambda_3 - \lambda_2} \left( e^{-\lambda_2 t} - e^{-\lambda_3 t} \right)$$

$$+ \frac{k_3}{k_1} N_1^0 \frac{\lambda_1}{\lambda_3 - \lambda_1} \left( \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right)$$  \hspace{1cm} (3.26)

Equations (3.22), (3.23), (3.24), (3.25) and (3.26) describe the "preferential release" mode.

If all species dissolve at the same fractional rate, then all have the same fractional rate constant $k$ ($k_i = k$), and Equation (3.22) becomes:

$$N_i(t) = \sum_{m=1}^{i} N_m^0 \left( \frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \sum_{n=1}^{i} \frac{e^{-(\lambda_n + k)t}}{\prod_{x=m}^{n} (\lambda_n - \lambda_x)}$$

where

$$N_m^0 = \frac{k M_m^0}{Q}$$  \hspace{1cm} (3.28)

The release mode of Equations (3.27) and (3.28) is called the "exponential release mode".

The release modes of Section 3.2 have been illustrated for the case of no diffusional transport of the released radionuclide at the point of release, so that a concentration boundary condition at the source can be specified, as in Equations (3.4) and (3.28). In the next
3.4 General Superposition Equation for Band Release (F1)

In this section, we develop the general superposition relation for the band release of decaying radionuclides through sorbing media. As shown before, there are two standpoints for connecting the nuclide-release phenomena with the nuclide migration. One is to treat the release as a boundary condition at the repository, and the transport equation without source term is solved using this boundary condition. The other is to treat the release as a source term in the transport equation.

With the first version, the transport equation (2.54) is reduced to

\[
L_i(N_i) \equiv -K_i \lambda_i N_i - \frac{\partial}{\partial z} \left( \frac{\partial N_i}{\partial z} \right) - \frac{\partial}{\partial z} \left( \varepsilon v A_i N_i \right) - \frac{\partial}{\partial t} \left( \varepsilon K_i N_i \right) - K_i \lambda_i N_i
\]

(3.34)

where \( N_i(z,t) \) is the nuclide concentration in water, \( v \) is the water velocity, \( D_i \) the effective diffusion coefficient and \( K_i \) is the overall retardation coefficient. \( A_i \) is defined in (2.55). For simplicity, the subscript \( f \) has not been included in (3.34). The governing equation for the "mother", i.e., the first member of the decay chain, is obtained
by formally setting \( \lambda_0 = 0 \). The properties \( \epsilon, D_i, \nu, A_i, B_i \) and \( K_i \) are considered generally to be functions of \( z \).

The initial condition is

\[
N_i(z,0) = 0 \quad , \quad i = 1,2,\ldots , \quad 0 < z < \infty \quad (3.35)
\]

We further prescribe

\[
N_i(z,t) = 0 \quad , \quad i = 1,2,\ldots , \quad 0 < z < \infty ; \quad t < 0 \quad (3.36)
\]

In the following we will be concerned with two special forms of the solution to the above equations. These are due to a step release of the radionuclides at the boundary \( z = 0 \), where they obey the conditions

\[
N_i^s(0,t) = B_i(t)h(t) \quad , \quad N_i^{*s}(0,t-T) = B_i(t)h(t-T) \quad (3.37)
\]

for \( i = 1,2,3\ldots n \). With the help of (3.37) we wish to construct the solution for the band release \( N_i^b(z,t) \) which satisfies the boundary conditions

\[
N_i^b(0,t) = B_i(t)h(t) - B_i(t)h(t-T) \quad (3.38)
\]

where \( T \) is the leach time for the band release. We will show that the desired solution is given by:

\[
N_i^b(z,t) = N_i^s(z,t) - N_i^{*s}(z,t-T) \quad , \quad i = 1,2,\ldots , \quad 0 < z < \infty ; \quad t > 0 \quad (3.39)
\]

where \( N_i^s(z,t) \) is the solution for the step release. Equation (3.37) and \( N_i^{*s}(z,t) \) is the step release solution satisfying the boundary condition:
\[ N^*_i(0,t) = B^*_i(t)h(t) = B_i(t+\tau)h(t) \]  

(3.40)

The system of equation (3.34) is linear in the \( N_i \) functions and simultaneously subject to non-trivial boundary conditions of the form of (3.37) for all of the nuclides. It is convenient to construct a solution through a set of subsidiary equations in which only one boundary condition arises, one at a time. Thus, we consider for example \( N^s_i(z,t) \) in \( 0 < z < \infty \), where \( i \) is any number of the chain \( 1 = 1, 2, \ldots, n \), with \( \lambda_o = 0 \). The contribution \( N^s_i(1) \) due to the finite initial amount of the first member of the chain is given by:

\[ L_i\left(N^s_i(1)\right) = -K_i - \lambda_i - \lambda_{i-1} N^s_i(1), \quad i \geq 1, \quad \lambda_o = 0 \]  

(3.41a)

\[ N^s_i(1)(0,t) = B_i(t)h(t), \quad N^s_i(1)(z,t < 0) = 0 \]  

(3.41b)

\[ N^s_i(1)(0,t) = 0, \quad (-\infty < t < \infty), \quad N^s_i(1)(z,t < 0) = 0, \quad i \geq 2 \]  

(3.41c)

The contribution \( N^s_i(2) \) due to the finite amount of the second member of the chain is given by:

\[ L_i\left(N^s_i(2)\right) = -K_i - \lambda_i - \lambda_{i-1} N^s_i(2), \quad \lambda_i = 0, \quad i \geq 2 \]  

(3.42a)

\[ N^s_i(2)(0,t) = B_i(t)h(t), \quad N^s_i(2)(z,t < 0) = 0 \]  

(3.42b)

\[ N^s_i(2)(0,t) = 0, \quad (-\infty < t < \infty), \quad N^s_i(2)(z,t < 0) = 0, \quad i \geq 3 \]  

(3.42c)

and so forth. In each subsequent subsystem, the starting index is increased by unity to represent the member which is to be a mother for the remainder of the chain. On account of the linearity it follows at once that
and that \(N_{i}^{s}(z,t)\) satisfied equations (3.34), (3.35), (3.36), (3.37) and (3.43). In exactly the same way one can construct the solution for the function \(N_{i}^{s}(z,t-T)\) by rewriting (3.41), (3.42) and (3.43) in terms of the argument \((t-T)\) after replacing \(B_{i}(t)\) by \(B_{i}^{*}(t)\). The combination of these two solutions giving \(N_{i}^{b}(z,t)\), as expressed in (3.39), satisfies the partial differential equation (3.34). It also satisfies the initial conditions because \(N_{i}^{s}(z,t)\) and \(N_{i}^{s}(z,t-T)\) vanish for \(t < 0\) and \(t < T\). The boundary conditions force a constraint between the function \(B_{i}(t)\) and \(B_{i}^{*}(t)\), which is described as

\[
B_{i}^{*}(t) = B_{i}(t+T) \tag{3.44}
\]

The second version of the nuclide release mode as mentioned previously asserts that the transport equation (3.34) is rewritten as

\[
L_{i}(N_{i}) = -\left(\gamma_{i_{-1}}^{l_{i-1}} + \gamma_{i_{-1}}^{f_{i}}(z,t)\right) + \epsilon_{i_{-1}}^{N_{i_{-1}}} - \epsilon_{i_{-1}}^{f_{i}}(z,t)
\]

\[
= -g_{i}(z,t), \quad i = 1, 2, \ldots \tag{3.45}
\]

where we postulate that nuclides are released into the water phase only.

At the boundary \(C\) of the defined domain \(D\), the following homogeneous condition is satisfied

\[
N_{i}(z,t) = 0, \quad z,t \leq C \tag{3.46}
\]
We introduce a function \( G_i(z,t;\xi,\tau) \) which satisfies

\[
L_i[G_i(z,t;\xi,\tau)] = -\delta(z-\xi) \delta(t-\tau) \tag{3.47}
\]

\( G_i(z,t;\xi,\tau) = 0 \), \( z,t \in \mathbb{C} \) \tag{3.48}

If we can find the solution for \( G_i(z,t;\xi,\tau) \), then \( N_i(z,t) \) is given by

\[
N_i(z,t) = \iint_D G_i(z,t;\xi,\tau) g_i(\xi,\tau) \, d\xi d\tau \tag{3.49}
\]

The above relation can be easily shown as follows: Multiplying both sides of (3.47) by \( g_i(\xi,\tau) \) and integrating the resultant equation over the domain \( D \), one can obtain

\[
\iint_D g_i(\xi,\tau) L_i[G_i(z,t;\xi,\tau)] \, d\xi d\tau = -\iint_D g_i(\xi,\tau) \delta(z-\xi) \cdot \delta(t-\tau) \, d\xi d\tau
\]

\[= -g_i(z,t) \tag{3.50}\]

The left hand side of (3.50) can be rewritten as

\[
\iint_D g_i(\xi,\tau) L_i[G_i(z,t;\xi,\tau)] \, d\xi d\tau = \iint_D L_i[G_i(z,t;\xi,\tau) g_i(\xi,\tau)] \, d\xi d\tau
\]

\[= L_i \left[ \iint_D G_i(z,t;\xi,\tau) g_i(\xi,\tau) \, d\xi d\tau \right] \tag{3.51}\]

Combining (3.50) and (3.51) and (3.45) we can find that (3.49) is the solution for (3.45) and (3.46). Equation (3.49) satisfies (3.45) because \( G_i(z,t;\xi,\tau) \) satisfies (3.48). Equation (3.49) can be rewritten as
\[ N_i(z,t) = \int \int_D G_i(z, t; \xi, \tau) \epsilon(\xi)f_i(\xi, \tau) d\xi d\tau + \int \int_D G_i(z, t; \xi, \tau) K_{i-1}(\xi) \epsilon(\xi) \lambda_{i-1} N_{i-1}(\xi, \tau) d\xi d\tau \]  

(3.52)

Equation (3.52) is a recursive solution. By iterative procedure, we can find that \( N_i(z,t) \) is given by the linear combination with respect to \( f_i, f_{i-1}, \ldots f_1 \). Then, the solution of the band release in which \( f_i^b \) is expressed by the combination of two step-release modes,

\[ f_i^b(z,t) = f_i^s(z,t)h(t) - f_i^s(z,t)h(t-T) \]  

(3.53)

is given by the linear combination of the solutions for these step releases:

\[ N_i^b(z,t) = N_i^s(z,t) - N_i^{s*}(z,t-T) \]  

(3.54)

\( N_i^{s*}(z,t) \) is the solution for the step release,

\[ f_i^s(z,t+T)h(t) = f_i^{s*}(z,t)h(t) \]  

(3.55)

Equation (3.54) agrees with (3.39) which has been obtained on account of the concentration boundary condition. When \( f_i^s(z,t) \) is expressed by

\[ f_i^s(z,t) = \phi_i(t) (z)h(t) ; \quad f_i^{s*}(z,t) = \phi_i^{s*}(t) \delta(z)h(t) \]  

(3.56)

the \( \phi_i \) term can be described with the help of (3.32) as follows:

\[ \phi_i(t) = QB_i(t)/S ; \quad \phi_i^{s*}(t) = QB_i^{s*}(t)/S \]  

(3.57)
Q is the volumetric flow rate of water and \( S \) is the effective cross section area of the water phase. Combining (3.57) with (3.55) we obtain

\[
B_i(t+T) = B_i^*(t) \tag{3.58}
\]

To illustrate how this condition is met, let us assume that the \( B_i(t) \) are given by Bateman equations with known coefficients \( B_{ij} \), which has been shown in Section 3.2. In this case, (3.44) can be expressed as

\[
B_i^*(t) = B_i(t+T) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} \tag{3.59}
\]

where \( B_{ij}^* = B_{ij} e^{-\lambda_j T} \)

Then, if we described the solution of the step release

\[
N_i^s(0,t) = B_i(t) h(t)
\]

or

\[
f_i^s(z,t) = \phi_i(t) \delta(z) (t)
\]

as \( N_i^s(z,t;B_{ij}) \), the solution for the band release can be expressed by:

\[
N_i^s(z,t) = N_i^s(z,t;B_{ij}) - N_i^s(z,t-T;B_{ij}^*) \tag{3.60}
\]

By use of the superposition equation, the relatively simple solutions for step releases can be used to construct the exact but more complicated solution for a band release.
Lester et al. (L1) have proposed the superposition relation for a band release

\[ N_i^b(z,t) = N_i^s(z,t; B_{ij}) - N_i^s(z,t-T; B_{ij}) \]  \hspace{1cm} (3.61)

Equation (3.61) is incorrect, because it does not take into account the difference between Bateman coefficients, \( B_{ij}^* \) and \( B_{ij} \). It is, in fact, applicable only to chemical chromatographic bands wherein radioactive decay does not occur. As seen from Equation (3.59), only when \( \lambda_i = 0 \) are the \( B_{ij}^* \) and \( B_{ij} \) equal. However, in applying Equation (3.61), exponential decay corrections were made to the subtractive term in the right-hand side to avoid the calculation of negative concentrations that would otherwise occur in some regions when radioactive decay occurs (B5). The correct superposition Equation (3.60) now shows the proper method of calculating band-release from concentrations for a multi-member decay chain.
4. RECURSIVE AND GENERAL SOLUTIONS OF THE TRANSPORT EQUATION FOR NUCLIDE MIGRATION

4.1 Introduction

We have obtained the fundamental transport equation (2.54) which governs the migration of the nuclide through geological media. When we neglect a term of \( f \) in (2.54), the governing equation can be expressed as

\[
\frac{\partial}{\partial t} (K_i \varepsilon N_i) + \frac{\partial}{\partial z} (A_i \varepsilon w N_i) - \frac{\partial}{\partial z} \left( \varepsilon D_i \frac{\partial N_i}{\partial z} \right) = \varepsilon (K_{i-1}^{\lambda_i - 1} N_{i-1} - K_i^{\lambda_i} N_i) + \varepsilon f_i(z,t) , \quad \lambda_0 = 0
\]  

(4.1)

\( K_i \) is the overall sorption retardation coefficient, \( \varepsilon \) is the porosity, \( v \), the water velocity, \( D_i \), the diffusion coefficient and \( A_i \) is given by (2.55). These values are functions of \( z \) and time in general.

When \( K_i, A_i, \varepsilon, v \) and \( D_i \) are independent of the distance \( z \) and time \( t \) the above equation can be simplified as

\[
\frac{\partial^2 N_i}{\partial z^2} - v \frac{\partial N_i}{\partial z} - K_i \frac{\partial N_i}{\partial z} - K_i^{\lambda_i} N_i = - K_{i-1}^{\lambda_i - 1} N_{i-1} - f_i(z,t) , \quad \lambda_0 = 0
\]  

(4.2)

In this chapter, we shall be concerned with solving the fundamental equation (4.2) with arbitrary release modes. Section 4.2 discusses the solution of the transport equation with a nuclide source. Section 4.3 treats the recursive solution for the transport equation without a source term, in which the release phenomena are treated as a boundary condition at \( z = 0 \). Section 4.4 is concerned with the case of no-dispersion. Recursive solutions of different forms and the general solution will be proposed for an \( i \)-member decay chain and arbitrary release modes.
Section 4.5 discusses the condition for local secular equilibrium in a decay chain of very rapidly decaying nuclides.

4.2 Solution of the Transport Equation with a Nuclide Source

4.2.1 Recursive solution of the transport

Equation (4.2) can be rewritten as

\[ \eta_i \frac{\partial^2 N_i}{\partial z^2} - v_i \frac{\partial N_i}{\partial z} - \frac{\partial N_i}{\partial t} - \lambda_i N_i = - \frac{K_{i-1}}{K_i} \lambda_i N_{i-1} - \frac{f_i(z,t)}{K_i} \]

\[ \equiv - f_i(z,t) \]

(4.3a)

\[ \eta_i \equiv \frac{D_i}{K_i} \quad v_i \equiv \frac{v}{K_i} \]

(4.3b)

The defined domain in which (4.3) hold is

\[ D : -\infty < z < \infty \quad , \quad -\infty < t < \infty \]

(4.4a)

and the concentrations \( N_i(z,t) \) at the boundary of the domain, \( C \), are zero:

\[ N_i(z,t) = 0 \quad , \quad z,t \in C \]

(4.4b)

The solution of (4.3) and (4.4) can be obtained with the help of the Green's function as shown in Section 3.4:

\[ N_i(z,t) = \iint_D G_i(z,t;\xi,\tau)f_i(\xi,\tau)d\xi d\tau \]

(4.5)

\( G_i(z,t;\xi,\tau) \) should satisfy
\[
\frac{\partial^2 G_i}{\partial z^2} - v_i \frac{\partial G_i}{\partial z} - \frac{\partial G_i}{\partial t} - \lambda_i G_i = -\delta(z-\xi)\delta(t-\tau) \tag{4.6a}
\]

\[
G_i(z,t;\xi,\tau) = 0 \quad z, t \in \mathbb{C} \tag{4.6b}
\]

The solution for \(G_i(z,t;\xi,\tau)\) can be obtained with the aid of the Fourier integral. The delta function is expressed by the Fourier integral as:

\[
\delta(z-\xi) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{ik(z-\xi)} \, dk, \quad \delta(t-\tau) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega(t-\tau)} \, d\omega \tag{4.7}
\]

Then, the right hand side of (4.6a) can be written as:

\[
-\delta(z-\xi)\delta(t-\tau) = \frac{1}{4\pi^2} \int_{-\infty}^{\infty} e^{ik(z-\xi)} \, dk \int_{-\infty}^{\infty} e^{-i\omega(t-\tau)} \, d\omega \tag{4.8}
\]

We prescribe the solution for \(G_i\) as:

\[
G_i(z,t;\xi,\tau) = \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{ik(z-\xi)-i\omega(t-\tau)} S(k,\omega) \, dk \, d\omega \tag{4.9}
\]

Introducing (4.9) into (4.6a), \(S(k,\omega)\) takes the form:

\[
S(k,\omega) = -\frac{1}{i\omega - (\eta_i k^2 + \lambda_i) - i\nu \, k} \tag{4.10}
\]

Then,

\[
G_i(z,t;\xi,\tau) = -\frac{1}{4\pi^2} \int_{-\infty}^{\infty} e^{ik(z-\xi)} I(k) \, dk \tag{4.11}
\]
where

\[
I(k) = \int_{-\infty}^{\infty} \frac{e^{-i\omega(t-\tau)}}{i\omega - (\eta_k^2 + \lambda_1) - iv_k} \, d\omega
\]  

(4.12)

Now we consider the integral \( I(k) \). The function in the integral is analytical except for

\[
\omega = -i(\eta_k^2 + \lambda_1) + v_k
\]  

(4.13)

Then, we can obtain \( I(k) \) in the time domain \( (t-\tau) < 0 \) as:

\[
I(k) = \lim_{R \to \infty} \int_{-R}^{R} \frac{e^{-i\omega(t-\tau)}}{i\omega - (\eta_k^2 + \lambda_1) - iv_k} \, d\omega = 0, \quad (t-\tau) < 0
\]  

(4.14)

because as indicated by the closed curve \( C_\rho \) in Fig. 4.1:

\[
\lim_{\rho \to \infty} \text{Re}(e^{-i\omega(t-\tau)}) \to 0 \quad \text{when} \quad (t-\tau) < 0
\]

Fig. 4.1. Domain of complex variable.
in the time domain \((t-\tau) > 0\), we can integrate along the closed curve, 
\(C_{\rho}\), because in this case,

\[
\lim_{\rho \to \infty} \text{Re}(e^{-i\omega(t-\tau)}) = 0 \quad \text{when } (t-\tau) > 0
\]

Then, the integral \(I(k)\) is given by:

\[
I(k) = -2\pi i [\text{Res.}]_{\omega} = -i(\lambda_i + \eta_i k^2) + v_i k
\]

\[
= -2\pi \exp[-(t-\tau)(\lambda_i + \eta_i k^2) - iv_i k(t-\tau)] \quad \text{, } (t-\tau) > 0 \quad (4.15)
\]

From (4.14) and (4.15), the integral is given by:

\[
I(k) = \begin{cases} 
0 & \text{, } (t-\tau) < 0 \\
-2\pi e^{-(t-\tau)(\lambda_i + \eta_i k^2) - iv_i k(t-\tau)} & \text{, } (t-\tau) > 0 
\end{cases} \quad (4.16)
\]

Introducing (4.16) into (4.11), \(G_i(z,t;\xi,\tau)\) is expressed as:

\[
G_i(z,t;\xi,\tau) = \begin{cases} 
\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp[i(k(z-\xi)-v_i(t-\tau)) - (\lambda_i + \eta_i k^2)(t-\tau)]dk & \text{, } (t-\tau) > 0 \\
0 & \text{, } (t-\tau) < 0 
\end{cases} \quad (4.17)
\]

For the integration of (4.17) we can use the formula:

\[
\int_{-\infty}^{\infty} e^{-a k^2 + b k} dk = \exp(b^2/4a) \sqrt{\pi/a} \quad , \quad a > 0 \quad (4.18)
\]

Then \(G_i(z,t;\xi,\tau)\) is explicitly obtained as:
\[ C_i(z,t;\xi,\tau) = \begin{cases} 
\frac{e^{-\lambda_i(t-\tau)}}{\sqrt{4\pi \eta_i(t-\tau)}} \exp \left[-\frac{(z-\xi-v_i(t-\tau))^2}{4\eta_i(t-\tau)}\right], & (t-\tau) > 0 \\
0, & (t-\tau) < 0 
\end{cases} \] 
(4.19)

The solution for (4.3) and (4.4) is obtained by introducing (4.19) into (4.5)

\[ N_i(z,t) = \int_{-\infty}^{t} \int_{-\infty}^{\infty} \left[ \frac{f_i(\xi,\tau)}{K_i} + \lambda_i \frac{K_{i-1}}{K_i} N_{i-1}(\xi,\tau) \right] d\xi d\tau \]
(4.20)

\[ x \frac{e^{-\lambda_i(t-\tau)}}{\sqrt{4\pi \eta_i(t-\tau)}} \exp \left[-\frac{(z-\xi-v_i(t-\tau))^2}{4\eta_i(t-\tau)}\right] d\xi d\tau \]

This is the general solution for arbitrary release modes.

When \( f_i(z,t) \) is expressed as:

\[ f_i(z,t)/K_i = \phi_i(z,t)h(t) \]
(4.21)

and \( N_{i-1}(z,t) \) is zero for time \( t < 0 \), then \( N_i(z,t) \) becomes

\[ N_i(z,t) = \int_{0}^{t} \int_{-\infty}^{\infty} \left[ \frac{f_i(\xi,\tau)}{K_i} + \lambda_i \frac{K_{i-1}}{K_i} N_{i-1}(\xi,\tau) \right] d\xi d\tau \]
(4.22)

\[ x \frac{e^{-\lambda_i(t-\tau)}}{\sqrt{4\pi \eta_i(t-\tau)}} \exp \left[-\frac{(z-\xi-v_i(t-\tau))^2}{4\eta_i(t-\tau)}\right] d\xi d\tau \]

Furthermore, we postulate that the nuclide is released at \( z = 0 \). In this case, \( f_i(z,t) \) is expressed by
\[ f_i(z,t)/K_i = \varphi_i(t)\delta(z)h(t) = f_i(t)\delta(z) \]  

(4.23)

Therefore, the nuclide concentration is reduced to a simplified form:

\[
N_i(z,t) = \int_0^t f_i(t-\tau) \frac{e^{-\lambda_i T}}{\sqrt{4\pi \eta_i T}} \exp \left[ -\frac{(z-v_i T)^2}{4\eta_i T} \right] d\tau 
\]

\[
+ \frac{\lambda_{i-1}K_{i-1}}{K_i} \int_0^t \int_0^\infty N_{i-1}(\xi,t-\tau) \frac{e^{-\lambda_i T}}{\sqrt{4\pi \eta_i T}} \exp \left[ -\frac{(z-v_i T-\xi)^2}{4\eta_i T} \right] d\xi d\tau
\]

(4.24)

The physical meaning of (4.24) is clear. The first term on the right hand side of (4.24) is the contribution of the \(i^{th}\) nuclide which is released directly from the repository. The amount of the \(i^{th}\) nuclide released from the repository at time \((t-\tau)\) during the time interval, \(d\tau\), is given by \(f_i(t-\tau)d\tau\). The probability density that the \(i^{th}\) nuclide released at time \((t-\tau)\) locates at the position \(z\), after time \(\tau\) has elapsed, is given by:

\[
\frac{1}{\sqrt{4\pi \eta_i T}} \exp \left[ -\frac{(z-v_i T)^2}{4\eta_i T} \right]
\]

(4.25)

The amount of the \(i^{th}\) nuclide also decreases by decay, so the amount of the nuclide which has been released at the time \((t-\tau)\) becomes \(f_i(t-\tau) \cdot \exp(-\lambda_i T)d\tau\) after the time interval, \(\tau\). Thus, the contribution of the \(i^{th}\) nuclide released from the repository is given by the first term on the right hand side of (4.24).
The second term of the right hand side of (4.24) arises from the 

\[ i \text{th} \] 
uclide generated from the \((i-1)\text{th}\) nuclide, which has been distrib-
uted through the whole region of the water path due to convection and
dispersion. The amount of the \(i\text{th}\) nuclide generated from the \((i-1)\text{th}\)
nuclide during the time interval, \(d\tau\), at the position, \(\xi\), and at the
time \((t-\tau)\), is given by:

\[
\frac{\lambda_{i-1} K_{i-1}}{K_i} N_{i-1}(\xi, t-\tau) d\tau
\]  

(4.26)

The probability that the \(i\text{th}\) nuclide generated in the range \(\xi\) to \((\xi + d\xi)\),
locates at the position, \(z\), during the time interval, \(\tau\), is given by:

\[
\frac{1}{\sqrt{4\pi \eta_i \tau}} \exp \left[ - \frac{(z-\xi-v_i \tau)^2}{4\eta_i \tau} \right] d\xi
\]

(4.27)

Thus the contribution of the \(i\text{th}\) nuclide generated from the \((i-1)\text{th}\)
nuclide for a time interval \(d\tau\) and a distance \(d\xi\) is given by the product
of (4.26) and (4.27).

When the dispersion coefficient becomes zero, the general solution
(4.22) yields the solution without dispersion, with the aid of the
relation:

\[
\lim_{\eta_i \to 0} \frac{1}{\sqrt{4\pi \eta_i \tau}} \exp \left[ - \frac{(\xi-v_i \tau)^2}{4\eta_i \tau} \right] = \delta(\xi-v_i \tau)
\]

(4.28)

The resultant solution is expressed by:
\[ N_i(z,t) = \int_0^t \frac{f_i(z-v_i \tau, t-\tau)}{K_i} e^{-\lambda_i \tau} d\tau \]

\[ + \int_0^t \frac{\lambda_i-1}{K_i} N_{i-1}(z-v_i \tau, t-\tau) e^{-\lambda_i \tau} d\tau \]

(4.29)

and if \( f_i(z,t) \) is given by (4.23), equation (4.29) is reduced to:

\[ N_i(z,t) = \frac{1}{v_i} \phi_i(t-z/v_i) e^{-z \lambda_i/v_i} h(t-z/v_i) \]

\[ + \int_0^{\text{Min}(t,z/v_i)} \frac{\lambda_i-1}{K_i} N_{i-1}(z-v_i \tau, t-\tau) e^{-\lambda_i \tau} d\tau \]

(4.30)

4.2.2 Concentration of the \( i^{\text{th}} \) nuclide in a decay chain

We can obtain the concentration of the \( i^{\text{th}} \) nuclide from the recursive formula, (4.24). From (4.3b) the following relations can be derived from setting \( D_i = D \)

\[ \eta_i = D v_i / v \equiv K v_i , \quad K_{i-1}/K_i = v_i/v_{i-1} \]

(4.31)

Introducing (4.31) into (4.24), one can obtain

\[ N_i(z,t) = \int_0^t f_i(t-\theta_i) e^{-\lambda_i \theta_i} \Phi(v_i \theta_i, z-v_i \theta_i) d\theta_i \]

(4.32)

\[ + \frac{\lambda_i-1}{v_{i-1}} \int_0^t \int_{-\infty}^{\infty} N_{i-1}(\xi, t-\theta_i) F(v_i \theta_i, z-\xi-v_i \theta_i) e^{-\lambda_i \theta_i} d\xi d\theta_i \]
where

\[ F(\alpha, \beta) = \frac{1}{\sqrt{4\pi K\alpha}} \exp \left( -\frac{\beta^2}{4K\alpha} \right) \]  \hspace{1cm} (4.33)

For the mother nuclide, \( i = 1 \), \( \lambda_0 = 0 \).

\[ N_1(z,t) = \int_{0}^{t} f_1(t-\theta)e^{-\lambda_1 \theta} F(v_1 \theta, z-v_1 \theta) d\theta \]  \hspace{1cm} (4.34)

With the help of (4.32), \( N_2(z,t) \) can be obtained as:

\[ N_2(z,t) = \int_{0}^{t} f_2(t-\theta_2)e^{-\lambda_2 \theta_2} F(v_2 \theta_2, z-v_2 \theta_2) d\theta_2 \]

\[ + \frac{\lambda_1 v_2}{v_1} \int_{0}^{t} \int_{0}^{t-\theta_2} f_1(t-\theta_1-\theta_2)e^{-\lambda_1 \theta_1-\lambda_2 \theta_2} \]

\[ \times \int_{-\infty}^{\infty} F(v_1 \theta_1, z-v_1 \theta_1) F(v_2 \theta_2, z-v_2 \theta_2) d\xi \]  \hspace{1cm} (4.35)

The integral term with respect to \( \xi \) is reduced to

\[ \int_{-\infty}^{\infty} F(a, \xi-a) F(b, z-\xi-b) d\xi = F(a+b, z-(a+b)) \]  \hspace{1cm} (4.36)

Using the above equation, \( N_2(z,t) \) can be rewritten by

\[ N_2(z,t) = \int_{0}^{t} f_2(t-\theta_2)e^{-\lambda_2 \theta_2} F(v_2 \theta_2, z-v_2 \theta_2) \]

\[ + \frac{\lambda_1 v_2}{v_1} \int_{0}^{t} \int_{0}^{t-\theta_2} f_1(t-\theta_1-\theta_2)e^{-\lambda_1 \theta_1-\lambda_2 \theta_2} \times F(v_1 \theta_1+v_2 \theta_2, z-(v_1 \theta_1+v_2 \theta_2)) \]  \hspace{1cm} (4.37)
Generally, the $N_i(z,t)$ is calculated by the same procedure.

\[
N_i(z,t) = \int_0^t \frac{f_i(t-\theta_i)e^{-\lambda_1^{\theta_i}F(\nu_i, \theta_{i}, z-\nu_i\theta_i)}}{0} d\theta_i \\
+ \sum_{j=1}^{i-1} \left( \frac{\nu_i}{\nu_j} \prod_{k=j}^{i-1} \lambda_k \right) \int_0^t d\theta_i \int_0^{c-\theta_i} d\theta_{i-1} \ldots \int_0^{t-\sum_{k=j+1}^{i} \theta_k} d\theta_j \quad (4.38)
\]

\[
x f_j(t-\sum_{k=j}^{i} \theta_k) e^{-\sum_{k=j}^{i} \lambda_k \theta_k} f\left( \sum_{k=j}^{i} \nu_k \theta_k, z-\sum_{k=j}^{i} \nu_k \theta_k \right)
\]

4.3 Recursive Solution of the Transport Equation with a

Concentration Boundary Condition

Consider the canonical system

\[
\begin{align*}
K_1 \frac{\partial N_1}{\partial t} + \nu \frac{\partial N_1}{\partial z} - D \frac{\partial^2 N_1}{\partial z^2} + \lambda_1^\theta_1 N_1 &= 0 \\
K_2 \frac{\partial N_2}{\partial t} + \nu \frac{\partial N_2}{\partial z} - D \frac{\partial^2 N_2}{\partial z^2} + \lambda_2^\theta_2 N_2 &= \lambda_1^\theta_1 N_1
\end{align*}
\]

\[(4.39)\]

\[
\begin{align*}
K_i \frac{\partial N_i}{\partial t} + \nu \frac{\partial N_i}{\partial z} - D \frac{\partial^2 N_i}{\partial z^2} + \lambda_{i-1}^\theta_{i-1} N_{i-1} &= \lambda_i^\theta_i N_i \\
\end{align*}
\]

which is to be solved for $N_i = N_i(z,t)$ in the domain $0 < z < \infty$, $o < t < \infty$ subject to the

initial condition: $N_i(z,0) = 0$, $z > 0$ \[(4.40)\]

boundary condition:

\[
-D^* \frac{\partial N_i}{\partial z} + \nu N_i = \nu N_i^0_{11}(t) \quad \text{for} \quad z = 0, \ t > 0 \quad (4.41)
\]
and \( N_i(z,t) \) and its derivatives must tend to zero with a suitable exponential order as \( z \to \infty \).

The boundary condition includes a contribution due to dispersion effects and if this term is not desired one can formally set \( D^* \) equal to zero in the final solution with certain additional changes which we indicate later. The boundary data \( \phi_i(t) \) are arbitrarily prescribed integrable functions.

It is convenient to introduce the following parameters:

\[
\alpha_i = \lambda_i K_i, \quad \beta_i = -\lambda_i - \gamma^2 n_i, \quad \gamma = v/2D, \quad n_i = D/K_i \quad (4.42)
\]

With the first of these one rewrites the general equation (4.39) as

\[
K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} - D \frac{\partial^2 N_i}{\partial z^2} + \alpha_i N_i = \alpha_{i-1} N_{i-1} \quad , \quad \alpha_0 = 0 \quad (4.43)
\]

We assume that \( N_{i-1}(z,t) \) is known and show how to compute \( N_i(z,t) \).

First one simplifies (4.43) by removing the convection and radioactive decay terms on the left hand side. For this let:

\[
N_i(z,t) = e^{\gamma z + \beta_i t} U_i(z,t) \quad (4.44)
\]

where \( U_i(z,t) \) is as yet an unknown function. Substitution of this into (4.43) results in the following partial differential equation for \( U_i \):

\[
\frac{\partial U_i}{\partial t} = \eta_i \frac{\partial^2 U_i}{\partial z^2} + S_{i-1}(z,t) \quad , \quad z > 0 \quad , \quad t > 0 \quad (4.45a)
\]
where

\[ S_{i-1}(z,t) = \frac{\alpha_{i-1}}{K_i} e^{-(\beta_i-\beta_{i-1})t} U_{i-1}(z,t) , \quad \alpha_0 = 0 \] (4.45b)

The initial and boundary conditions, (4.40) and (4.41), transform into:

\[ U_{i}(z,0) = 0 , \quad z > 0 \] (4.46)

and

\[ \frac{\partial U_{i}}{\partial z} - \gamma U_{i} = -2N_{i} \gamma g_{i}(t) , \quad \text{at} \quad z = 0 , \quad t > 0 \] (4.47a)

where

\[ g_{i}(t) = e^{-\beta_{i}t} \phi_{i}(t) \] (4.47b)

We exclude in the following the cases when \( \gamma = 0 \) and \( \eta_i = 0 \). In view of (4.44), \( U_{i} \) and its derivatives must tend to zero faster than \( e^{-\gamma z} \) as \( z \to \infty \). The governing equations of the problem have thus been reduced to (4.45) - (4.47). We solve this problem in two different forms.

4.3.1 Integral transform method

One defines a modified Fourier transform and its inverse with help of:

\[ u_{i}(p,t) = \int_{0}^{\infty} T(p,z) U_{i}(z,t) dz \] (4.48)

and

\[ U_{i}(z,t) = \int_{0}^{\infty} T(p,z) u_{i}(p,t) dp \] (4.49)
This transform kernel has the form (Cl)

\[ T(p,z) = \sqrt{\frac{2}{\pi}} \frac{pcos(pz) + \gamma sin(pz)}{\sqrt{p^2 + \gamma^2}} \]  \hspace{1cm} (4.50)

and satisfies the Sturm-Liouville system:

\[ \frac{d^2 T}{dz^2} + p^2 T = 0 \hspace{0.5cm} , \hspace{0.5cm} 0 < z < \infty \]  \hspace{1cm} (4.51a)

\[ \frac{dT}{dz} - \gamma T = 0 \hspace{0.5cm} , \hspace{0.5cm} at \ z = 0 \]  \hspace{1cm} (4.51b)

If one applies the integral transform (4.48) to equation (4.45) there results:

\[ \frac{d}{dt} u_i(p,t) = \eta_i \int_0^\infty \left[ \frac{\partial^2 u_i}{\partial z^2} \cdot T(p,z)dz + S_{i-1}(p,t) \right] \]  \hspace{1cm} (4.52a)

where

\[ S_{i-1}(p,t) = \int_0^\infty S_{i-1}(z,t)T(p,z)dz \]  \hspace{1cm} (4.52b)

By integration by parts one obtains with the help of equations (4.48) and (4.51a)

\[ \int_0^\infty \frac{\partial^2 u_i}{\partial z^2} \cdot T(p,z)dz = \left[ T \frac{\partial u_i}{\partial z} - u_i \frac{\partial T}{\partial z} \right]_0^\infty - p^2 u_i(p,t) \]  \hspace{1cm} (4.53)
The bracketed term vanishes at the upper limit in view of the fact that $U_i$ and its derivatives must vanish faster than $e^{-\gamma z}$ as $z \to \infty$. At the lower limit $z = 0$, there results with the help of (4.47) and (4.51),

$$
- \left[ T \frac{3 U_i}{3 z} - U_i \frac{2 T}{3 z^2} \right]_{z=0} = T(p,0)\gamma [2\eta_1^0 g_i(t) - U_i(0,t)] + U_i(0,t)\gamma T(p,0)
$$

$$
= 2T(p,0)\eta_1^0 g_i(t)
$$

(4.54)

If one combines (4.53) and (4.54) and substitutes the result into (4.52) one obtains the inhomogeneous Bateman equations

$$
\frac{d u_i(p,t)}{dt} + \eta_1^0 p^2 u_i(p,t) = r_{i-1,i}(p,t)
$$

(4.55a)

where

$$
r_{i-1,i}(p,t) = S_{i-1}(p,t) + T(p,0)2\gamma_1^0 N_i^0 g_i(t)
$$

(4.55b)

with $S_0(p,t) = 0$

The source term $R_{i-1,i}(z,t)$ consists of the transform of the precursor solution and the boundary condition of the $i$th nuclide. Equation (4.55) is solved subject to the transformed initial conditions (4.46) i.e.,

$$
u_i(p,0)=0,
$$

$$
u_i(p,t) = \int_0^t e^{-\eta_1^0 p^2(t-\tau)} r_{i-1,i}(p,\tau) d\tau
$$

(4.56)

This can finally be inverted with the help of (4.49) and yields the formal solution for $U_i(z,t)$, or in terms of (4.44), $N_i(z,t)$:
\[ N_i(z,t) = e^{\gamma z + \beta_i t} \int_0^\infty e^{-\eta_i p^2 t} T(p,z) \left\{ \int_0^t e^{\eta_i p^2 \tau} r_{i-1,i}(p,\tau) d\tau \right\} dp \]

(4.57)

The source term \( r_{i-1,i} \) has the form:

\[ r_{i-1,i}(p,\tau) = \int_0^\infty S_{i-1}(z,\tau) T(p,z) dz + 2\gamma T(p,0) \eta_i N_i^0 g_i(\tau) \]

(4.58)

with

\[ S_o(z,t) \equiv 0 \]

One can separate the solution into the two basic contributions which stem from release at the boundary of the \( i^{th} \) nuclide and the radioactive decay of its precursors

\[ N_i(z,t) = e^{\gamma z + \beta_i t} \left[ \int_0^\infty e^{-\eta_i p^2 t} T(p,0) T(p,z) \left\{ \int_0^t e^{\eta_i p^2 \tau} g_i(\tau) d\tau \right\} dp \right. \]

\[ \left. + \int_0^\infty e^{-\eta_i p^2 t} T(p,z) \left\{ \int_0^t e^{\eta_i p^2 \tau} \left[ \int_0^\infty S_{i-1}(z,\tau) T(p,z) dz \right] d\tau \right\} dp \right] \]

(4.59)

Consider now the special case that the diffusive component at the boundary vanishes (i.e., \( D^* = 0 \)) in (4.41). Then one can use a standard Fourier sine transform and the kernel in equation (4.50) is replaced by:
\[ T(p, z; D^* = 0) = \sqrt{\frac{2}{\pi}} \sin(pz) \quad (4.60) \]

The term \( 2\gamma T(p, 0) \) in Eq. (4.58) has been replaced by \( dT(p, 0)/dz \). The final solution reads

\[
N_1(z, t) = e^{Yz+B*_{11}t} \left[ \eta_1 N_1^1 \int_0^\infty e^{-\eta_1 p^2} T(p, z; D^* = 0) \frac{dT(p, 0; D^* = 0)}{dz} \right]
\]

\[
\left[ \int_0^t e^{\eta_1 p^2} g_1(t) dt \right] dp + \int_0^\infty e^{\eta_1 p^2} T(p, z; D^* = 0) \left[ \int_0^t e^{\eta_1 p^2} \right]
\]

\[
\left( \int_0^\infty (s_{i-1}(z, \tau) T(p, z; D^* = 0) dz) dt \right) \left[ \int_0^t e^{\eta_1 p^2} \right]
\]

\[ \left. \right] \left. \right] \left. \right] \left. \right] \left. \right] , \quad s_0(t, z) = 0 \quad (4.61) \]

4.3.2 The Green's function method

Let us replace the variables \( z \) and \( t \) by \( \xi \) and \( \tau \) respectively in the solution \( U_i(z, t) \) of the equation (4.45) i.e.,

\[
\frac{\partial U_i}{\partial t} = \eta_i \frac{\partial^2 U_i}{\partial z^2} + S_{i-1}(z, t) , \quad z > 0, \ t > 0 \quad (4.62) \]

Let \( G(z, \xi, \tau) \) be the Green's function of the homogeneous Equation (4.62) which for boundary condition of type III, (i.e., (4.47)) has the form (Cl) given below:
\begin{align*}
G(z, \xi, \tau) &= \frac{1}{\sqrt{4\pi \eta_1 \tau}} \left\{ \left[ e^{-\frac{(z-\xi)^2}{4\eta_1 \tau}} + e^{-\frac{(z+\xi)^2}{4\eta_1 \tau}} \right] - 2\gamma \int_0^\infty e^{-\frac{(z+\xi+z')^2}{4\eta_1 \tau}} \frac{dz'}{d(z')^2} \right\} \\
0 < z, \xi < \infty, z \neq \xi, \tau > 0 \\
\end{align*}
(4.63)

Now replace in $G$ the argument $\tau$ by $(t-\tau)$. Then $G$ satisfies the equation

\begin{equation}
-\frac{\partial G}{\partial \tau} = \eta_i \frac{\partial^2 G}{\partial \xi^2}, \quad \xi > 0, \quad \tau > 0 \\
(4.64)
\end{equation}

and the homogeneous boundary condition

\begin{equation}
\frac{\partial G}{\partial \xi} - \gamma G = 0, \quad \xi = 0 \\
(4.65)
\end{equation}

Using now (4.62) and (4.64) one has:

\begin{equation}
-\frac{\partial (GU_i)}{\partial \tau} = G \frac{\partial U_i}{\partial \tau} + U_i \frac{\partial G}{\partial \tau} = \eta_i \left[ G \frac{\partial^2 U_i}{\partial \xi^2} - U_i \frac{\partial^2 G}{\partial \xi^2} \right] + GS_{i-1} \\
(4.66)
\end{equation}

If one integrates this with respect to $\xi$ from 0 to $\infty$ and then with respect to $\tau$ from 0 to $(t-\theta)$, where $0 < \theta < t$, one obtains:

\begin{align*}
\int_0^\infty \left[ C \frac{\partial U_i}{\partial \xi^2} - U_i \frac{\partial^2 G}{\partial \xi^2} \right] d\xi + \int_0^\infty GS_{i-1} d\xi \\
= \eta_i \int_0^{t-\theta} dt \int_0^\infty \left[ C \frac{\partial U_i}{\partial \xi^2} - U_i \frac{\partial^2 G}{\partial \xi^2} \right] d\xi + \int_0^{t-\theta} dt \int_0^\infty GS_{i-1} d\xi \\
(4.67)
\end{align*}
If the first integral on the right hand side is integrated by parts this yields:

\[
\int_{0}^{\infty} (GU_i)_{\tau=t-\theta} d\xi = \int_{0}^{\infty} (GU_i)_{\tau=0} d\xi + \eta_i \int_{0}^{t-\theta} \left( \frac{\partial U_i}{\partial \xi} \right) \bigg|_{\xi=0}^{\xi=\infty} - \left( U_i \frac{\partial G}{\partial \xi} \right) \bigg|_{\xi=0}^{\xi=\infty}
\]

\[
- \int_{0}^{\infty} \left[ \frac{\partial G}{\partial \xi} \frac{\partial U_i}{\partial \xi} - \frac{\partial U_i}{\partial \xi} \frac{\partial G}{\partial \xi} \right] d\xi \bigg|_{\xi=0}^{\xi=\infty} + \int_{0}^{t-\theta} \int_{0}^{\infty} G_{i-1} d\xi d\tau
\]

(4.68)

On the right hand side the first term vanishes on account of the initial condition, (4.46), and so does the third integral. The second integral vanishes at the upper limit \( \xi = \infty \) for the reasons stated after (4.47) about the behavior of \( U_i \). At the lower limit

\[
\left( \frac{\partial G}{\partial \xi} \frac{\partial U_i}{\partial \xi} - \frac{\partial U_i}{\partial \xi} \frac{\partial G}{\partial \xi} \right) \bigg|_{\xi=0} = G \left[ \gamma U_i - 2\gamma N_{i-1} \gamma g_i(t) \right] \bigg|_{\xi=0} - \left( U_i \gamma G \right) \bigg|_{\xi=0}
\]

\[
= - G(z,0,t-\tau)2\gamma N_{i-1} g_i(t)
\]

(4.69)

on account of (4.47) and (4.65). Hence,

\[
\int_{0}^{\infty} G(z,\xi,\theta)U_i(\xi,t-\theta) d\xi = 2\gamma N_{i-1} \int_{0}^{t-\theta} G(z,0,t-\tau) g_i(\tau) d\tau
\]

\[
+ \int_{0}^{t} d\tau \int_{0}^{\infty} G(z,\xi,t-\tau) S_{i-1}(\xi,\tau) d\xi
\]

(4.70)
We now pass to the limit $\theta \to 0$ and recall the delta-function character of the Green's function $G(z, \xi, \theta)$ when $\theta \to 0$. The integral on the left hand side sifts out the value of $U_i$ at $(z, t)$ so that the solution to the problem is given by:

$$
U_i(z, t) = 2\gamma \eta_1 N_1^0 \int_{0}^{t} G(z, 0, t-\tau) g_i(\tau) d\tau 
$$

$$
+ \int_{0}^{t} d\tau \int_{0}^{\infty} G(z, \xi, t-\tau) s_{i-1}(\xi, \tau) d\xi
$$

(4.71)

where $G$ is defined by (4.63).

In the case when the dispersion term in the boundary condition (4.41) or its equivalent (4.47) is absent $G$ must satisfy a boundary condition of type I, i.e., in place of equations (4.65) we have

$$
G = 0, \quad \xi = 0.
$$

(4.72)

The proper form of $G$ is then

$$
G(z, \xi, \tau) = \frac{1}{\sqrt{4\pi \eta_1 \tau}} \left\{ e^{-\frac{(z-\xi)^2}{4\eta_1 \tau}} - e^{-\frac{(z+\xi)^2}{4\eta_1 \tau}} \right\},
$$

(4.73)

$$
0 < z < \infty, \quad 0 < \xi < \infty, \quad z \neq \xi, \quad \tau > 0
$$

and (4.71) takes on the form
\[
U_i(z,t) = \eta_i N_1^o \int_0^t \frac{\partial G(z,o,t-\tau)}{\partial \xi} s_i(\tau) d\tau \\
+ \int_0^t dt \int_0^\infty G(z,\xi,t-\tau) S_{i-1}(\xi,\tau) d\xi
\]

(4.74)

4.4 Recursive and General Solutions of the Transport Equation

without Dispersion

4.4.1 Recursive solution of the transport equation without dispersion and some properties of the solution

Consider the canonical system

\[
K_1 \frac{\partial N_1}{\partial t} + v \frac{\partial N_1}{\partial z} + \lambda_1 K_1 N_1 = 0
\]

\[
K_2 \frac{\partial N_2}{\partial t} + v \frac{\partial N_2}{\partial z} + \lambda_2 K_2 N_2 = \lambda_1 K_1 N_1
\]

\[
\vdots \quad \vdots \quad \vdots \quad \vdots \\
\vdots \quad \vdots \quad \vdots \quad \vdots \\
\vdots \quad \vdots \quad \vdots \quad \vdots \\
K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} + \lambda_i K_i N_i = \lambda_{i-1} K_{i-1} N_{i-1}
\]

(4.75)

which is to be solved for \( N_i \approx N_i(z,t) \) in the domain \( 0 < z < \infty \), \( 0 < t < \infty \) subject to the side conditions that

\[
N_i(o,t) = N_i^o \phi_i(t), \; t > 0, \text{ with } \phi_i(t) \equiv 0, \; t < 0 \quad (4.76a)
\]

\[
N_i(z,0) = 0, \; z > 0 \quad (4.76b)
\]
The boundary data \( \phi_i(t) \) are arbitrary integrable functions which describe the release of the members of the chain from the repository.

The general form of the recursive equation system (4.75) is

\[
\frac{\partial N_i}{\partial t} + v_o \frac{\partial N_i}{\partial z} + \lambda_i N_i = \frac{K_{i-1}}{K_i} \lambda_{i-1} N_{i-1} \quad \text{with} \quad \lambda_0 = 0 \tag{4.77}
\]

where

\[
v_i = v/K_i
\]

We assume that \( N_{i-1}(z,t) \) is known and show how to compute \( N_i(z,t) \).

We construct the solution in two different forms, each of which exhibits certain advantages in demonstrating certain physical properties of the solution.

A. **Integral Transform Method**

Consider the Laplace transform of \( N_i(z,t) \) with respect to \( z \) and define

\[
n_i(p,t) = \int_0^\infty e^{-pz} N_i(z,t) \, dz
\]

\[
n_{i-1}(p,t) = \int_0^\infty e^{-pz} N_{i-1}(z,t) \, dz
\]

The application of the transform operator to the equation (4.77) and use of equation (4.76a) yields:

\[
\frac{dn_i(p,t)}{dt} + \left( pv_i + \lambda_i \right) n_i(p,t) = \frac{K_{i-1}}{K_i} \lambda_{i-1} n_{i-1}(p,t) + v_i N_i^0 \phi_i(t)
\]

\[
(4.79)
\]
This ordinary differential equation is solved subject to the initial condition \( n_i(p,0) = 0 \) which results from the application of equation (4.76b). The solution is

\[
n_i(p,t) = \int_0^t \left\{ \frac{K_i - 1}{K_i} \lambda_i^{-1} n_{i-1}(p,t-\tau) + v_i N_i \phi_i(t-\tau) \right\} e^{-(p v_i + \lambda_i)\tau} d\tau
\]

(4.80)

To invert \( n_i(p,t) \) we make use of the "shift" rule and apply it to the first term on the right hand side.

\[
\mathcal{L}^{-1} \left\{ \int_0^t \frac{K_i - 1}{K_i} \lambda_i^{-1} n_{i-1}(p,t-\tau)e^{-p v_i \tau} e^{-\lambda_i \tau} d\tau \right\}
\]

\[
= \frac{K_i - 1}{K_i} \lambda_i^{-1} \int_0^t e^{-\lambda_i \tau} K_i n_{i-1}(z - v_i \tau, t-\tau) d\tau
\]

provided \( v_i > 0 \).

The second term in (4.80) is seen to be a Laplace transform, if, as stated above, \( \phi_i(t) \equiv 0 \) for \( t < 0 \). Then:
\[ v_1 N_1^0 \int_0^t \phi_i(t-\tau) e^{-p v_1 \tau} e^{-\lambda_1 \tau} d\tau = v_1 N_1^0 \int_0^\infty e^{-p v_1 \tau} e^{-\lambda_1 \tau} \phi_i(t-\tau) d\tau \]

\[ = N_1^0 \int_0^\infty e^{-p v_1 \tau} e^{-\lambda_1 z} \phi_i \left( t - \frac{z}{v_1} \right) dz \]

This shows that the second contribution in (4.80) has the inverse:

\[ \mathcal{L}^{-1} \left\{ v_1 N_1^0 \int_0^t \phi_i(t-\tau) e^{-p v_1 \lambda_1 \tau} d\tau \right\} = N_1^0 e^{-\lambda_1 z} \phi_i \left( t - \frac{z}{v_1} \right) \]

Thus the complete inverse of (4.80) is

\[ N_1(z,t) = N_1^0 e^{-\lambda_1 z} \phi_i \left( t - \frac{z}{v_1} \right) + \frac{K_{i-1}}{K_1} \lambda_{i-1} \int_0^t e^{-\lambda_1 \tau} N_{i-1}(z-v_1 \tau, t-\tau) d\tau \]

(4.81)

Now the precursor concentration \( N_{i-1}(z,t) \equiv 0 \) for \( t < 0, z < 0 \). Hence, the upper limit in the integral can be changed to \( \text{Min}(t, z/v_1) \) so that the final result reads:

\[ N_1(z,t) = N_1^0 e^{-\lambda_1 z} \phi_i \left( t - \frac{z}{v_1} \right) + \frac{K_{i-1}}{K_1} \lambda_{i-1} \int_0^{\text{Min}(t, z/v_1)} e^{-\lambda_1 \tau} N_{i-1}(z-v_1 \tau, t-\tau) d\tau \]

(4.82)
This equation yields the concentration of the \( i^{th} \) member of the chain in terms of the \((i-1)^{th}\) member and can hence be used in a recursive manner. Starting with the mother concentration \( N_1(z,t) \) solved from the first of the equations (4.75) one computes from (4.82), \( N_2(z,t) \) etc.

The physical interpretation of the solution is simple. The first term on the right hand side represents those \( i^{th} \) nuclides that have been convected from the boundary \((z = 0)\) and have escaped decay (the exponential attenuation term) while propagating along the characteristic \((t-z/v_1) = \) constant. The second term represents those \( i^{th} \) nuclides which are contributed from the \((i-1)^{th}\) nuclides lying along the characteristic \((z-v_{i_1}t) = \) constant with their exponential attenuation.

\[ N_i(z,t) = \sum_{r=1}^{i} e^{-\frac{\lambda_i z}{v_r}} \phi_{i_1,1}(t-z/v_1) \]

\[ (4.84) \]

B. Classical Method

We observe from (4.82) that the equation \( N_1(z,t) \) has the simple solution

\[ N_1(z,t) = N_1^o e^{-\lambda_1 z/v_1} \phi_1(t-z/v_1) \]

\[ (4.83) \]

This can be verified by direct substitution into (4.76) and (4.77). This in turn suggests that we try for the \( i^{th} \) nuclide concentration the generalization

\[ N_i(z,t) = \sum_{r=1}^{i} e^{-\frac{\lambda_i z}{v_r}} \phi_{i_1,r}(t-z/v_r) \]

\[ (4.84) \]
The functions $N_{i,r}(t-z/v_r)$ are at this point unknown and are to be found. For $i = 1$, (4.84) reduces to:

$$N_1(z,v_1) = e^{-\frac{\lambda}{v_1}z} \mathcal{K}_{1,1}(t-z/v_1)$$  \hspace{1cm} (4.85)

which identifies

$$N_{1,1}(t-z/v_1) = \mathcal{K}_{1,1}^0(t-z/v_1)$$  \hspace{1cm} (4.86)

To find the functions $N_{i,r}(t-z/v_r)$ lets substitute the proposed solution form (4.84) into (4.77) with $N_{i,r} = dN_{i,r}/d(t-z/v_r)$.

$$\sum_{r=1}^i e^{-\frac{\lambda_r}{v_r}z} \left[ 1 - \frac{v_i}{v_r} \right] N_{i,r}^r \left( t - \frac{z}{v_r} \right) + \sum_{r=1}^i e^{-\frac{\lambda_r}{v_r}z} \left[ \lambda_i - \frac{v_i}{v_r} \lambda_r \right] N_{i,r} \left( t - \frac{z}{v_r} \right)$$

$$= \frac{K_{i-1}}{K_i} \sum_{r=1}^{i-1} e^{-\frac{\lambda_r}{v_r}z} N_{i-1,r} \left( t - \frac{z}{v_r} \right)$$  \hspace{1cm} (4.87)

For $r = i$, the terms of the left hand side of (4.87) vanish so with $t-z/v_r = t$. For $r < i-1$ there results on equating the coefficients of the exponential terms,

$$\left[ \frac{1}{v_i} - \frac{1}{v_r} \right] N_{i,r}^r(t) + \left[ \frac{\lambda_i}{v_i} - \frac{\lambda_r}{v_r} \right] N_{i,r}^r(t) = \frac{K_{i-1} \lambda_i - 1}{v} N_{i-1,r}(t)$$  \hspace{1cm} \text{for } r = 1,2,\ldots,(i-1)  \hspace{1cm} (4.88)$$
The initial condition (4.76b) is satisfied in view of (4.84) if one chooses

\[ N_{i,r}(\tau) = 0 \quad , \quad \tau \leq 0 \quad (4.89) \]

The boundary condition (4.76a) is satisfied if

\[ \sum_{r=1}^{i} N_{i,r}(\tau) = N_{i-1,1}(\tau) \quad (4.90) \]

when

\[ t > 0 \quad \text{and} \quad z = 0 \]

We now solve the equation system (4.88) - (4.90) recursively. Let

\[ \frac{\lambda_i}{\nu_i} - \frac{\lambda_r}{\nu_r} \]

\[ \frac{1}{\nu_i} - \frac{1}{\nu_r} = \Delta_{i,r} \quad (4.91) \]

then (4.88) can be written as:

\[ \left( \frac{1}{\nu_i} - \frac{1}{\nu_r} \right) \frac{d}{d\tau} \begin{bmatrix} \Delta_{i,r} N_{i,r}(\tau) \end{bmatrix} = \lambda_{i-1} \frac{K_{i-1}}{\nu} \begin{bmatrix} \Delta_{i,r} N_{i-1,r}(\tau) \end{bmatrix} \]

(4.92)

which has the solution subject to the initial condition (4.89):

\[ N_{i,r}(\tau) = \frac{\lambda_{i-1} K_{i-1}}{K_i - K_r} e^{-\Delta_{i,r} \tau} \int_{0}^{\tau} N_{i-1,r}(\tau')e^{\Delta_{i,r} \tau'} d\tau' , \]

\[ r = 1,2,\ldots,(i-1) \]

(4.93)
The remaining member \( N_{i,1}(t) \) is then calculated from (4.84) and (4.90) as
\[
N_{i,1}(t) = N_{i,1}^0 \phi_1(t) - \sum_{r=1}^{i-1} N_{i,r}(t)
\] (4.94)

(4.93) and (4.94) determine the \( N_{i,r} \) functions recursively from those of \( N_{i-1,r} \) and with these the solution for \( N_1(z,t) \) is completed. The variables \( \tau \) and \( t \) in these equations are dummy variables. In order to keep the conditions in equation (4.89) clearly in mind, one must multiply the \( N_{i,r} \) functions by unit step functions. The final solution then takes on the form
\[
N_1(z,t) = \sum_{r=1}^{i-1} \left[ e^{-\frac{\lambda_z}{v_r} z} N_{i,r}(t - \frac{z}{v_r}) h\left(t - \frac{z}{v_r}\right) - e^{-\frac{\lambda_1}{v_1} z} \right] N_{i,r}(t - \frac{z}{v_1}) h\left(t - \frac{z}{v_1}\right) + N_{i,1}^0 \phi_1(t - \frac{z}{v_1})
\] (4.95)

The physical interpretation of the solution is identical to that discussed in the previous method given in Section A. The first term (sum) represents those \( i^{th} \) nuclides which are contributed from all nuclides \( r = 1,2,\ldots,(i-1) \) with their exponential attenuation while the last term represents those \( i^{th} \) nuclides that have been convected from the boundary and have escaped decay.

An important additional feature of the solution (4.95) is that it allows one to find the range in the \( t \) and \( z \) variables in which the solution is non-trivial. For this, consider the first term on the right
hand side of (4.95). The term in the bracket vanishes when both step functions vanish. This will occur for a time value smaller than the smallest of the values, \( z/v_k \), \( k = 1,2,\ldots,f,\ldots i \). Suppose this smallest value occurs for the migration velocity \( v_f \), then \( z/v_f \) will represent the first arrival time of the nuclides at position \( z \). If furthermore, \( v_f > v_i \), the second term on the right hand side of (4.95) is also zero. Consequently, we have

\[
N_1(z,t) = 0 \quad \text{for} \quad t \leq z/v_f
\]  

(4.96)

This can be represented graphically at a fixed \( z \) position as shown by Figure 4.2.

![Graph](image)

Figure 4.2. Concentration versus time at fixed position.

On the other hand, at a fixed time the above result can be written as

\[
N_1(z,t) = 0 \quad , \quad z \geq v_f t
\]  

(4.97)
Physically \( z < v_f t \) defines the region that has been contaminated at time \( t \) and \( z = v_f t \) represents the leading edge of the contaminant wave. This is illustrated by Fig. 4.3.

![Diagram](image)

**Figure 4.3.** Concentration profile against distance at fixed time.

Furthermore, the rate at which \( N_i(z,t) \) rises (from zero) at the first arrival time at \( z = \text{constant} \) or at the leading wave front edge at \( t = \text{constant} \) is controlled by the term

\[
N_i(t - \frac{z}{v_f}) e^{-\left(\frac{\lambda_f}{v_f}\right)z} h(t - \frac{z}{v_f})
\]

The numerical calculations for a three-member chain shown in the later chapter support all of the above conclusions.

In the case of an impulse release at \( z = 0 \) one can, in addition, also determine the cut-off in the arrival time (at fixed \( z \)) and the trailing wave front edge (at constant \( t \)) of the contaminant wave quite easily. The solution for the mother concentration is, in view of (4.85) and (4.86):
\[ N_1(z,t) = N_1^0 e^{-\left(\frac{\lambda_1}{v_1}\right)z} \delta(t - z/v_1) \]  

(4.98)

With \( N_{1,1} \) given by (4.86), (4.93) yields:

\[ N_{2,1}(\tau) = N_1^0 \frac{\lambda_1 K_1 T}{K_2 - K_1} e^{-\Delta_{12} \tau} \]  

(4.99)

so that

\[ N_2(z,t) = N_1^0 \frac{\lambda_1 K_1 T}{K_2 - K_1} e^{-\left(\frac{\lambda_1 - \lambda_2}{v_1 - v_2}\right)z - \Delta_{12} t} \]

\[ \times \left\{ h\left(t - \frac{z}{v_1}\right) - h\left(t - \frac{z}{v_2}\right) \right\} + N_2^0 e^{-\left(\frac{\lambda_2}{v_2}\right)z} \delta\left(t - \frac{z}{v_2}\right) \]  

(4.100)

In a comparable way, one can write down at once the expression for \( N_3(z,t) \).

Since the bracketed term divided by \((K_2 - K_1)\) is always positive, the contribution to \( N_2 \) from the mother will be non-trivial for

i) \( v_2 > v_1 \): \( v_1 t < z < v_2 t \)  

(4.101)

ii) \( v_2 < v_1 \): \( v_2 t < z < v_1 t \)
Outside this range \( N_2(z,t) \equiv 0 \). The edges of the wave fronts are immediately determined from (4.101) depending on the relative magnitude of \( v_1 \) and \( v_2 \).

Finally we note that the wave front slopes are also completely determined in terms of the exponential attenuation factors.

4.4.2 The general solution of the transport equations without dispersion

Consider the canonical system

\[
\begin{align*}
K_1 \frac{\partial N_1}{\partial t} + v \frac{\partial N_1}{\partial z} + \lambda_1 K_1 N_1 &= 0 \\
K_2 \frac{\partial N_2}{\partial t} + v \frac{\partial N_2}{\partial z} + \lambda_2 K_2 N_2 &= \lambda_1 K_1 N_1 \\
&\vdots \\
K_i \frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} + \lambda_i K_i N_i &= \lambda_{i-1} K_{i-1} N_{i-1} \\
&\vdots
\end{align*}
\]

(4.102)

which is to be solved for \( N_i = N_i(z,t) \) in the domain \( 0 < z < \infty \), \( 0 < t < \infty \) subject to the side conditions that

\[
N_i(0,t) = N^0_i \phi_i(t), \quad t > 0 \quad \text{with} \quad \phi_i(t) \equiv 0, \quad t < 0 \quad (4.103a)
\]

\[
N_i(z,0) = 0, \quad z > 0 \quad (4.103b)
\]
The boundary data $\phi_i(t)$ are arbitrary integrable functions which describe the release of the members of the chain from the repository.

The general form of the equation system (4.102) is

$$\frac{\partial N_i}{\partial t} + v_i \frac{\partial N_i}{\partial z} + \lambda_i N_i = \frac{K_{i-1}}{K_i} \lambda_{i-1} N_{i-1}, \quad \text{with} \quad \lambda_0 = 0$$

(4.104)

where

$$v_i = v/K_i$$

The aim is to obtain the general (i.e., non-recursive) solution for $N_i(z,t)$. For this purpose, consider the following $i$-sets of boundary conditions.

\begin{align*}
\text{Boundary Condition} & \quad \ell = 1 \quad \ell = 2 \quad \ldots \quad \ell = \ell \quad \ldots \quad \ell = i \\
\hline
j = 1 & N_{1}^{(1)}(0,t) = N_{1}^0 \phi_1(t) \quad N_{2}^{(1)}(0,t) = 0 \quad \ldots \quad N_{\ell}^{(1)}(0,t) = 0 \quad \ldots \quad N_{i}^{(1)}(0,t) = 0 \\
\vdots & \quad \vdots \quad \quad \vdots \quad \quad \vdots \quad \quad \vdots \\
\ell = \ell & N_{1}^{(\ell)}(0,t) = 0 \quad N_{2}^{(\ell)}(0,t) = N_{2}^0 \phi_2(t) \quad N_{\ell}^{(\ell)}(0,t) = N_{\ell}^0 \phi_\ell(t) \quad N_{i}^{(\ell)}(0,t) = 0 \\
\vdots & \quad \vdots \quad \quad \vdots \quad \quad \vdots \\
\text{Boundary Condition} & \quad \ell = 1 \quad \ell = 2 \quad \ldots \quad \ell = \ell \quad \ldots \quad \ell = i \\
\hline
j = 1 & N_{1}^{(1)}(0,t) = 0 \quad N_{2}^{(1)}(0,t) = 0 \quad \ldots \quad N_{\ell}^{(1)}(0,t) = 0 \quad \ldots \quad N_{i}^{(1)}(0,t) = N_{i}^0 \phi_i(t)
\end{align*}

(4.105)
For a given \( i^{th} \)-member of a chain we solve (4.103b) and (4.104) for each precursor \( \ell = 1, 2, \ldots, i \) subject to each set of boundary conditions on a line \( j \). Solutions satisfying these boundary conditions are denoted by \( N_\ell^{(j)}(x,t) \) \( \ell = 1, 2, \ldots, i \). This is repeated for every line \( j = 1, 2, \ldots, i \). We will produce a total number of \( i^2 \) solutions \( N_\ell^{(j)}(z,t) \).

On account of the linearity of (4.104) the solutions for the concentration of an \( i \)-member chain will be given by:

\[
N_i(z,t) = N_i^{(1)}(z,t) + \sum_{j=1}^{i-1} N_i^{(j)}(z,t) + \sum_{j=1}^{i} N_i^{(j)}(z,t)
\]

(4.106a)

For the \( i^{th} \) member the general form can be written as:

\[
N_i(z,t) = N_i^{(i)}(z,t) + \sum_{j=1}^{i-1} N_i^{(j)}(z,t)
\]

(4.106b)

According to (4.106b), in order to obtain the concentration of the \( i^{th} \) member, every solution \( N_i^{(j)}(z,t) \), \( j = 1, 2, \ldots, i \) must be specified.

For a given \( j \) the function \( N_i^{(j)}(z,t) \) depends intrinsically on its precursors solutions \( N_\ell^{(j)}(z,t) \) (i.e., \( \ell = 1, 2, \ldots i \)).
Therefore, in order to explicitly express \( N_\lambda^{(j)}(z,t) \) one must obtain first of all the precursor solutions. For these, the following equation (4.107a) must be solved for each \( N_\lambda^{(j)}(z,t) \), \( \lambda = 1,2,...i \) for a given \( j \).

\[
\frac{\partial N_\lambda^{(j)}}{\partial t} + \nu \frac{\partial N_\lambda^{(j)}}{\partial z} + \lambda \frac{\partial N_\lambda^{(j)}}{\partial \lambda} = \frac{K_{\lambda-1}}{K_\lambda} \lambda_{\lambda-1} N_{\lambda-1}^{(j)} , \ \lambda_0 = 0
\]

(4.107a)

After this procedure is repeated for every \( j = 1,2,...i \) one will have obtained all the \( N_\lambda^{(j)}(z,t) \) in equation (4.106b) and therefore the concentration \( N_i \) of the \( i \)th member is obtained.

The reader should note that some of the \( N_\lambda^{(j)}(z,t) \) functions are identically equal to zero due to their boundary conditions. Consider for instance the line \( j = 2 \) in (4.105) where \( N_1^{(2)}(0,t) = 0 \) but \( N_2^{(2)}(0,t) \neq 0 \). Then \( N_1^{(2)}(z,t) \equiv 0 \) for \( z > 0, \ t > 0 \). Analogously if \( N_1^{(3)}(0,t) \) and \( N_2^{(3)}(0,t) \) are both zero then \( N_1^{(3)}(z,t) \equiv N_2^{(3)}(z,t) \equiv 0 \) for \( z > 0, \ t > 0 \). In general there will be no precursors for the functions \( N_\lambda^{(j)}(z,t) \), which arise from the boundary conditions lying below the diagonal in (4.105),

\[
N_\lambda^{(j)}(z,t) \equiv 0 , \ \text{for} \ \lambda \leq j , \ z > 0 , \ t > 0
\]

(4.107b)

The boundary conditions (4.105) can be expressed in a more compact form as:

\[
N_\lambda^{(j)}(0,t) = \begin{cases} 
0 & \lambda \neq j \\
N_j \phi_j(t) & \lambda = j , \ t > 0 , \ (\phi_j(t) \equiv 0 , \ t < 0) 
\end{cases}
\]

(4.107c)

For a fixed \( j \) the boundary conditions which lie on the diagonal in (4.105) give rise to a set of chain members \( N_\lambda^{(j)}(z,t) \) for \( \lambda \geq j \).
The initial condition is

\[ N_{\ell}^{(j)}(z,0) = 0 \quad , \quad z > 0 \quad , \quad \ell > j \]  \hspace{1cm} (4.107d)

The reader can verify that the solutions to (4.107) when substituted into (4.106b) will satisfy the original equation system (4.103) and (4.104).

The functions \( N_{\ell}^{(j)}(z,t) \) are obtained by a repeated Laplace transform with respect to \( t \) and \( z \) of (4.107 a,b). Let

\[ n_{\ell}^{(j)}(z,s) = \int_{0}^{\infty} e^{-st} N_{\ell}^{(j)}(z,t) \, dt \quad , \quad \phi_{j}(s) = \int_{0}^{\infty} e^{-st} \phi_{j}(t) \]  \hspace{1cm} (4.108)

\[ n_{\ell}^{(j)}(p,s) = \int_{0}^{\infty} e^{-pz} N_{\ell}^{(j)}(z,s) \, dz \]  \hspace{1cm} (4.109)

There results from (4.107 a,b,c,d)

\[ p_{\ell}^{(j)}(p,s) - N_{\ell}^{0} \phi_{j}(s) + \mu_{\ell} n_{\ell}^{(j)}(p,s) = \nu_{\ell-1} n_{\ell-1}^{(j)}(p,s) \]

\[ j = 1, 2, \ldots i, \quad \ell > j \]  \hspace{1cm} (4.110a)

\[ n_{\ell-1}^{(j)}(p,s) \equiv 0 \quad , \quad j = 1, 2, \ldots i, \quad \ell \leq j \]  \hspace{1cm} (4.110b)

where

\[ \mu_{\ell} = \frac{s + \lambda_{\ell}}{\nu_{\ell}} \quad , \quad \nu_{\ell-1} = \frac{\lambda_{\ell-1}}{\nu_{\ell-1}} \quad , \quad \lambda_{0} = 0 \]  \hspace{1cm} (4.110c)
Equation (4.107c) implies that

\[
N_\ell^0 = \begin{cases} 
0, \ell \neq j \\
N_j^0, \ell = j 
\end{cases}
\]  

(4.110d)

We solve (4.110a) in a recursive manner from \( \ell = 1 \) to \( \ell = j \) for each value of \( j \); \( j \) runs from \( j = 1 \) to \( j = i \) since we are interested in the \( i \)th nuclide of a given chain.

Starting with \( j = 1 \) and letting \( \ell \) run through the values, 1, 2, ..., \( i \) one takes from (4.110b) \( n_0^{(1)}(p,s) = 0 \) and from (4.110d) \( N_2^0 = N_3^0 = \cdots = N_i^0 = 0 \). Therefore, (4.110a) yields in succession

\[
n_1^{(1)}(p,s) = \frac{N_1^{0\phi_1}(s)}{p + \mu_1} \\
n_2^{(1)}(p,s) = \frac{v_1 n_1^{(1)}(p,s)}{p + \mu_2} = \frac{v_1 N_1^{0\phi_1}(s)}{(p+\mu_1)(p+\mu_2)} \\
\vdots \\
n_i^{(1)}(p,s) = \frac{v_1 v_2 \cdots v_{i-1} N_1^{0\phi_1}(s)}{(p+\mu_1)(p+\mu_2) \cdots (p+\mu_{i-1})}
\]

(4.111)

Next one takes \( j = 2 \) and lets \( \ell \) run through the values, 2, ..., \( i \). From (4.110b) and (4.110d) one has \( n_1^{(2)}(p,s) = 0 \) and \( N_2^0 \neq 0 \), \( N_3^0 = N_4^0 = \cdots N_i^0 = 0 \) respectively. Hence (4.110a) yields
Continuing in this manner one shows that in general

\[ n^{(2)}_1(p,s) = \frac{N_2^0 \phi^0_2(s)}{p + \mu_2} \]

\[ n^{(2)}_2(p,s) = \frac{\nu_2 n^{(2)}_1(p,s)}{p + \mu_3} = \frac{\nu_2 N_2^0 \phi^0_2(s)}{(p + \mu_2)(p + \mu_3)} \]

\[ \vdots \]

\[ n^{(2)}_i(p,s) = \frac{\nu_2 \nu_3 \cdots \nu_{i-1} N_2^0 \phi^0_2(s)}{(p+\mu_2)(p+\mu_3)\cdots(p+\mu_i)} \]

(4.112)

Continuing in this manner one shows that in general

\[ n^{(j)}_i(p,s) = \frac{A^{(j)}_i N^0_j \phi_j(s)}{\prod_{m=j}^{i-1} (p + \mu_m)} \quad , \quad i > j \]

(4.113)

where

\[ A^{(j)}_i = \prod_{r=j}^{i-1} \nu_r = \prod_{r=j}^{i-1} \frac{\lambda_r}{\nu_r} \]

(4.113a)

while for \( j = i \) one has:

\[ n^{(i)}_i(p,s) = \frac{N^0_i \phi_i(s)}{p + \mu_i} \]

(4.114)

It can be verified after the inversion that the solutions \( N^{(j)}_1(z,t) \) do indeed satisfy the boundary conditions in (4.105) for \( i \geq j \).

By partial fraction expansion one can rewrite (4.113) as

\[ n^{(j)}_i(p,s) = A^{(j)}_i N^0_j \phi_j(s) \sum_{m=j}^{i} \frac{C^j_m(s)}{p + \mu_m} \quad , \quad i > j \]

(4.115)

provided that the \( \mu_m \) are all unequal. Here
\[
C_m^{(j)}(s) = \left[ \prod_{r=j}^{i} \left( \mu_r - \mu_m \right) \right]^{-1} 
\]

(4.116)

On inversion of (4.114) and (4.115) with respect to \( z \) one obtains

\[
n_i^{(i)}(z,s) = N_i^0 e^{-\mu_i z} \phi_i(s) \]

(4.117)

\[
n_i^{(j)}(z,s) = A_i^{(j)} N_j^0 \phi_j(s) \sum_{m=j}^{i} C_m^{(j)}(s) e^{-\mu_m z} \]

(4.118)

Since

\[
\mu_r - \mu_m = \Gamma_{rm} (s + \Delta_{rm}) 
\]

(4.119)

where

\[
\Gamma_{rm} = \frac{v_m - v_r}{\nu_m \nu_r}, \quad \Delta_{rm} = \frac{\lambda_r v_m - \lambda_m v_r}{\nu_m - \nu_r} 
\]

(4.118) becomes with the help of (4.112), (4.116) and (4.119)

\[
n_i^{(j)}(z,s) = A_i^{(j)} N_j^0 \sum_{m=j}^{i} \phi_j(s) e^{-\left( \frac{s + \nu_m}{\nu_m} \right) z} \prod_{r=j}^{i} \Gamma_{rm} \prod_{r=m}^{i} (s + \Delta_{rm}) 
\]

(4.120)
We transform the right hand side prior to inversion with respect to \( t \). Let

\[
\prod_{\substack{r=j \\ r\neq m}}^{i} G_{rm} = B_{m}^{(j)}, \quad \frac{1}{\prod_{\substack{r=j \\ r\neq m}}^{i} (s+\Delta_{rm})} = \sum_{\substack{r=j \\ r\neq m}}^{i} \frac{D_{rm}^{(j)}}{s + \Delta_{rm}}
\]  

(4.121)

where by partial fraction expansion

\[
D_{rm}^{(j)} = \left[ \prod_{\substack{q=j \\ q\neq m \neq r}}^{i} (\Delta_{qm} - \Delta_{rm}) \right]^{-1}
\]  

(4.122)

(i-1)

where \( D_{r,m} \) = \( i \).

With these (4.120) reads:

\[
r_{i}^{(j)}(z,s) = A_{i}^{(j)} \sum_{m=j}^{N} \frac{e^{-(\lambda_{m}/v_{m})z}}{B_{m}^{(j)}} \sum_{\substack{r=j \\ r\neq m}}^{i} D_{rm}^{(j)} \phi_{i}(s) e^{-\frac{(z/v_{m})s}{s + \Delta_{rm}}}
\]  

(4.123)

By the shift rule

\[
\mathcal{L}^{-1} \begin{pmatrix}
-(z/v_{m})s \\
\frac{e}{s + \Delta_{rm}}
\end{pmatrix} = g_{rm}^{(j)}(t) = \begin{cases}
0, & t < \frac{z}{v_{m}} \\
\Delta_{rm}(t - \frac{z}{v_{m}}), & t > \frac{z}{v_{m}}
\end{cases}
\]  

(4.124)
so that by the convolution rule

\[
N_i^{(j)}(z,t) = A_i^{(j)} N_j^0 \sum_{m=j}^{i} \frac{e^{-(\lambda_i^m/v_i^m)z}}{B_{m,m}^{(j)}} \sum_{\substack{r=0 \atop r \neq m}}^{i} D_{rm}^{(j)} (g_{rm}(t) \otimes \phi_j(t))
\]  

(4.125a)

where

\[
g_{rm}(t) \otimes \phi_j(t) = \int_0^t g_{rm}(\tau) \phi_j(t-\tau) d\tau
\]

(4.125b)

In view of the restriction placed on \( \phi_i(t) \) in (4.103), the inverse of (4.117) is

\[
N_i^{(i)}(z,t) = N_i^0 e^{-(\lambda_i/v_i)z} \phi_i(t-z/v_i)
\]

(4.126)

The substitution of (4.125) and (4.126) into (4.106) yields the general (non-recursive) solution
\[ N_i(z,t) = N_{i1}^0 e^{-\frac{\lambda_i}{v_i} z} \phi_i(t - z/v_i) \]

\[
+ \sum_{j=1}^{i-1} A_{ij}^0 \sum_{m=j}^{i} e^{-\frac{\lambda_m}{v_m} z} \sum_{r=j}^{i} D_{rm}^j (g_{rm}(t)\delta_{j}(t)) \sum_{r=m} \frac{D_{rm}^j}{B_m(j)} \sum_{r=j}^{i} D_{rm}^j (g_{rm}(t)\delta_{j}(t)) \]

(4.127)

Again the first term represents those \( i^{th} \) nuclides that have been
convected from the boundary \( (z = 0) \) and have escaped decay. The triple
sum represents those \( i^{th} \) nuclides contributed from all precursors.

For instance, when \( i = 1 \) the summations in equation (4.127) vanish
and the concentration for the 1st member is:

\[ N_1(z,t) = N_{11}^0 e^{-\frac{\lambda_1}{v_1} z} \phi_1(t - z/v_1) \]  

(4.127a)

For the second member of a chain, one sets \( i = 2 \) in (4.127)
resulting in

\[ N_2(z,t) = N_{21}^0 e^{-\frac{\lambda_2}{v_2} z} \phi_2(t - z/v_2) + \sum_{j=1}^{1} A_{2j}^0 \sum_{m=j}^{2} e^{-\frac{\lambda_m}{v_m} z} \sum_{r=j}^{2} D_{rm}^j (g_{rm}(t)\delta_{j}(t)) \sum_{r=m} \frac{D_{rm}^j}{B_m(j)} \sum_{r=j}^{2} D_{rm}^j (g_{rm}(t)\delta_{j}(t)) \]

The first summation has only one term for \( j = 1 \); the second
summation goes from \( m = j = 1 \) to \( m = 2 \) while the third summation will
have only one term for each value of \( m \) because \( r \neq m \). Expanding these
sums results in:
\[ N_2(z, t) = N_2^0 e^{-\left(\frac{\lambda_2}{v_2}\right) z} \phi_2(t - z/v_2) + A_2^{(1)} \nu_1^0 \left\{ e^{-\left(\frac{\lambda_1}{v_1}\right) z} \frac{d_{12}^{(1)}}{b_1^{(1)}} \left[ \phi_{21}(t) e^{q_1(t)} \right] \right\} \]

\[ + \frac{e^{-\left(\frac{\lambda_2}{v_2}\right) z}}{b_2^{(1)}} \ D_{12}^{(1)} \left[ \phi_{12}(t) e^{q_1(t)} \right] \]

\[ (4.127b) \]

By using (4.121) one has:

\[ B_1^{(1)} = \prod_{r=1}^{2} \frac{\Gamma_{r1}}{\Gamma_{21}} = \frac{v_1 - v_2}{v_1 v_2} \]

\[ B_2^{(1)} = \prod_{r=1}^{2} \frac{\Gamma_{r2}}{\Gamma_{12}} = \frac{v_2 - v_1}{v_1 v_2} \]

\[ (4.127c) \]

And using (4.122) one obtains:

\[ D_{z1}^{(1)} = D_{12}^{(1)} = 1 \]

\[ (4.127d) \]

From (4.124)

\[ g_{21}(t) = e^{-\Delta_{21}(t-z/v_1)} h(t-z/v_1) \]

\[ g_{12}(t) = e^{-\Delta_{12}(t-z/v_2)} h(t-z/v_2) \]

\[ (4.127e) \]

where \( h(t-z/v_m) \) is the unit step function.

Finally from (4.110c) and (4.113a)

\[ A_2^{(1)} = \frac{\lambda_1}{v_1} \]

\[ (4.127f) \]
Substituting (4.127c), (4.127d), (4.127e) and (4.127f) back into (4.127b) yields:

\[
N_2(z,t) = N_2^0 e^{-(\lambda_2/\nu_2) z} \phi_2(t-z/\nu_2) + \frac{N_2^0 \nu_2 \lambda_1}{\nu_1 - \nu_2} \left\{ e^{-(\lambda_1/\nu_1) z} \left[ e^{-\Delta_{21}(t-z/\nu_1)} h(t-z/\nu_1) \phi_2(t) \right] \right. \\
- e^{-(\lambda_2/\nu_2) z} \left[ e^{-\Delta_{12}(t-z/\nu_2)} h(t-z/\nu_2) \phi_2(t) \right]
\]

(4.127g)

The above equation is the solution for the 2nd member for an arbitrary boundary condition.

Let us consider the simple case of an impulse release at the boundary. This release is mathematically represented by:

\[
\phi_1(t) = \delta(t) T 
\]

(4.127h)

where \( \delta(t) \) is the Dirac's delta function.

By performing the convolution integral in (4.127g) using (4.127h)

\[
N_2(z,t) = N_2^0 e^{-(\lambda_2/\nu_2) z} \delta(t-z/\nu_2) + \frac{N_2^0 \lambda_1}{\nu_2 - \nu_1} \exp \left( -\Delta_{21} t - \frac{\lambda_2 - \lambda_1}{\nu_2 - \nu_1} z \right) \left[ h(t-z/\nu_1) - h(t-z/\nu_2) \right]
\]

(4.127i)

The above solution agrees with the previously obtained solution equation (4.100).

Similar developments will yield the solution for any further member of a radionuclide chain.
4.5 Local Secular Equilibrium in a Porous Medium

There are some decay chains in which very rapidly decaying nuclides are involved. For example, an important decay chain is

\[
{\begin{array}{cccc}
237_{\text{Np}} & \rightarrow & 233_{\text{Pa}} & \rightarrow & 233_{\text{U}} & \rightarrow & 229_{\text{Th}} & \rightarrow & 225_{\text{Ra}} \\
2.14 \times 10^6 \text{y} & & 27 \text{d} & & 1.62 \times 10^5 \text{y} & & 7340 \text{y} & & 15 \text{d}
\end{array}}
\]

(4.128)

For this chain in a stationary system we know that each of the short-lived nuclides e.g., \(^{233}\text{Pa}\) and \(^{225}\text{Ra}\), can reach secular equilibrium with its immediate precursor. In this section, we will determine the criteria for reaching secular equilibrium when radionuclide migration occurs.

The solution of the nuclide migration equation in recursive form is given by (4.32):

\[
N_i(z,t) = \int_0^t f_i(t-\theta) e^{-\lambda_i \theta} F(v_i \theta, z-v_i \theta) d\theta
\]

\[+ \frac{\lambda_{i-1} v_i}{v_{i-1}} \int_0^t \int_{-\infty}^{\infty} N_{i-1}(\xi, t-\theta) F(v_i \theta, z-\xi-v_i \theta) e^{-\lambda_i \theta} d\xi d\theta
\]

where

\[
F(v_i \theta, z-\xi-v_i \theta) = \frac{1}{\sqrt{4 \pi v_i \theta}} \exp \left[ - \frac{(z-\xi-v_i \theta)^2}{4 v_i \theta} \right]
\]
Let us consider the case such that the decay constant for the $i$th nuclide is much larger than that for the $(i-1)$th nuclide:

$$\lambda_i \gg \lambda_{i-1}$$  \hspace{1cm} (4.29)

The time interval, $t$, of interest here is of the order $1/\lambda_{i-1}$, thus the following equation is satisfied:

$$\frac{1}{\lambda_i} \ll t \approx \text{order of } 0(1/\lambda_{i-1}) = 0(1/\lambda_{i-1})$$  \hspace{1cm} (4.130)

Because $\lambda_i \gg 1$, the value of $\exp(-\lambda_i \theta_i)$ in (4.32) is nearly zero except for $\theta_i$ in the range of: $0 < \theta_i < 0(1/\lambda_i)$. This is illustrated in Figure 4.4. The function, $F(v_i \theta_i, z-\xi - v_i \theta_i)$, in (4.32) is approximated by a delta function, $\delta(z-\xi)$. This approximation is valid under the

![Diagram](XBL_817-1031)

**Figure 4.4.** Illustrative behavior of $F$ for local secular equilibrium.
conditions that $AB \gg BC$ and $4\kappa v_{1\perp} \overline{OA} \ll z^2$, where point A is a location of order $(1/\lambda_{1\perp})$ at $\xi = 0$ and point B is at $(z, \overline{OA})$. C is a point on the straight line, $(z - v_{1\perp} \theta_{1\perp}) = \xi$, at $\theta_{1\perp} = \overline{OA}$. The effect of the above conditions means that $F(v_{1\perp} \theta_{1\perp}, z - \xi - v_{1\perp} \theta_{1\perp})$ can be approximated by a delta function at the point C. Since $\overline{OA}$ is of the order $(1/\lambda_{1\perp})$, $AB = z$ and $BC = v_{1\perp} \overline{OA}$, the above two conditions are rewritten as:

\[ z \gg v_{1\perp}/\lambda_{1\perp}, \quad 4\kappa v_{1\perp}/\lambda_{1\perp} \ll z^2 \]  

(4.131)

Under the conditions of (4.131), the second term in (4.32) is given by:

\[ \frac{v_{1\perp} \lambda_{1\perp - 1}}{v_{1\perp - 1}} \int_0^t \int_{-\infty}^{\infty} N_{1-1}(\xi, t - \theta_{1\perp}) e^{-\lambda_{1\perp} \theta_{1\perp}} F(v_{1\perp} \theta_{1\perp}, z - \xi - v_{1\perp} \theta_{1\perp}) d\xi d\theta_{1\perp} \]

\[ \approx \frac{v_{1\perp} \lambda_{1\perp - 1}}{v_{1\perp - 1}} \int_0^t N_{1-1}(z, t - \theta_{1\perp}) e^{-\lambda_{1\perp} \theta_{1\perp}} d\theta_{1\perp} \]

\[ \approx \frac{v_{1\perp} \lambda_{1\perp - 1}}{v_{1\perp - 1}} N_{1-1}(z, t) \int_0^\infty e^{-\lambda_{1\perp} \theta_{1\perp}} d\theta_{1\perp} = \frac{v_{1\perp} \lambda_{1\perp - 1}}{v_{1\perp - 1} \lambda_{1\perp}} N_{1-1}(z, t) \]

(4.132)

In the above derivation, we have used the condition that $N_{1-1}(z, t)$ is almost constant in the time interval $\theta_{1\perp}$ of the order $1/\lambda_{1\perp}$. With the conditions of (4.131) the first term in the right hand side of (4.32) can also be reduced to:

\[ \int_0^t f_{1\perp}(t - \theta_{1\perp}) e^{-\lambda_{1\perp} \theta_{1\perp}} F(v_{1\perp} \theta_{1\perp}, z - v_{1\perp} \theta_{1\perp}) d\theta_{1\perp} = \delta(z) \int_0^t e^{-\lambda_{1\perp} \theta_{1\perp}} f_{1\perp}(t - \theta_{1\perp}) d\theta_{1\perp} \]

(4.133)
because \( P(\nu_i \theta_i, z-\nu_i \theta_i) \) is approximated as \( \delta(z) \). Then the first term can be neglected at distance \( z > 0 \).

From (4.132) and (4.133), we can obtain

\[
\frac{\lambda_i N_i(z,t)}{v_i} = \frac{\lambda_{i-1} N_{i-1}(z,t)}{v_{i-1}} \tag{4.134a}
\]

or

\[
\lambda_i N_i(z,t) = \lambda_{i-1} K_{i-1} N_{i-1}(z,t) \tag{4.134b}
\]

Equation (4.134b) shows that the local secular equilibrium is attained between the \( i^{th} \) and the \( (i-1)^{th} \) nuclides. The necessary conditions are summarized as:

\[
\lambda_i \gg \lambda_{i-1}, \quad z \gg v_i/\lambda_i, \quad \frac{\lambda_i z^2}{v_i} \gg 4\kappa, \quad t\lambda_i \gg 1
\]

(4.135)

\( N_{i+1}(z,t) \) is obtained from (4.32) as:

\[
N_{i+1}(z,t) = \int_0^t f_{i+1}(t-\theta_{i+1}) \frac{-\lambda_i \theta_{i+1}}{v_{i+1}} F(\nu_{i+1} \theta_{i+1}, z-\nu_{i+1} \theta_{i+1}) d\theta_{i+1}
\]

\[
+ \frac{\lambda_i v_{i+1}}{v_1} \int_0^t \int_{-\infty}^0 N_1(\xi, t-\theta_{i+1}) \frac{-\lambda_{i+1} \theta_{i+1}}{v_{i+1}} F(\nu_{i+1} \theta_{i+1}, z-\xi-\nu_{i+1} \theta_{i+1}) \xi d\xi d\theta_{i+1} \tag{4.136}
\]

Introduction of (4.134b) into (4.136) yields:

\[
N_{i+1}(z,t) = \int_0^t f_{i+1}(t-\theta_{i+1}) \frac{-\lambda_i \theta_{i+1}}{v_{i+1}} F(\nu_{i+1} \theta_{i+1}, z-\nu_{i+1} \theta_{i+1}) d\theta_{i+1}
\]

\[
+ \frac{v_i + \lambda_i}{v_{i-1}} \int_0^t \int_{-\infty}^0 d\theta_{i+1} \int_{-\infty}^0 dN_{i-1}(\xi, t-\theta_{i+1}) \frac{-\lambda_{i+1} \theta_{i+1}}{v_{i+1}} F(\nu_{i+1} \theta_{i+1}, z-\xi-\nu_{i+1} \theta_{i+1}) \tag{4.137}
\]
Equation (4.137) shows that, under the conditions of (4.135), nuclide \( i \) reaches secular equilibrium, and the \( n \)-member decay chain \( (\ldots, i-1, i, i+1, \ldots, n) \) reduces to the \((n-1)\) member decay chain \( (\ldots, i-1, i+1, \ldots, n) \).
5. APPLICATION OF GENERAL SOLUTIONS OF THE TRANSPORT EQUATION

5.1 Introduction

In the preceding chapters, we have discussed the different release modes and have obtained general solutions of the transport equation for arbitrary release modes.

In this chapter, explicit solutions of the space-time-dependent concentration of a radionuclide for various release modes will be developed. In Section 5.2 the general solution governing the one-dimensional dispersion-free migration of an i-member decay chain will be shown for several release modes. In Sections 5.3 and 5.4, the solutions for a three-member decay chain with dispersion are shown for the step and the band release modes. In Sections 5.5 and 5.6 we present numerical results based on these solutions. The migration features of some important decay chains are discussed in detail.

5.2 Application of the General Solution of the Transport Equation

Without Dispersion—Solutions for Several Release Modes

In the preceding chapter, the general solution of the transport equation without dispersion was shown in an explicit form:

\[ N_i(z,t) = e^{-\lambda_i z/v_i} N_i(t-z/v_i) \]

\[ + \sum_{\ell=1}^{i-1} \left( \prod_{r=\ell}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=\ell}^{i-1} e^{-\lambda_m z/v_m} \prod_{r=\ell}^{i} \frac{\Gamma_r}{\Gamma_{rm}} \sum_{r=\ell}^{m-1} D_{rm} \left[ \Delta_{rm}(t) \otimes N_\ell(t) \right] \]  

(5.1)
where
\[ g_{rm}(t) = \begin{cases} 
0, & t < \frac{z}{v_m} \\
-\Delta_{rm}(t-z/v_m), & t \geq \frac{z}{v_m}
\end{cases} \]

In this section, we present the solutions for several release modes with the help of (5.1), utilizing the properties of these release modes discussed in Section 3.2.

5.2.1 Solution for preferential release

The preferential release mode has been defined in Section 3.2 and is given by

\[ N_i^p(0,t) = N_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\frac{\Lambda_j t}{\lambda_j}} h(t) \] (3.22)

\[ B_{ij} = \sum_{m=1}^{j} \frac{\left( \frac{K_i}{K_m} \right) N_m^0 \frac{1}{\lambda_i} \prod_{r=m} \lambda_r}{\prod_{s=m}^{i} \left( \lambda_s - \Lambda_j \right)} \] (3.23)

The concentration of the \(i^{th}\) member can be obtained by introducing (3.22) into (5.1)
\[
N_i^0(z,t) = e^{-\lambda x/z_{v_i}} \sum_{j=1}^{i-1} B_{ij} \sum_{m=\ell}^{i} e^{-\lambda x/z_{m}} h(t-z/v_m) + \sum_{\ell=1}^{i-1} \left( \prod_{r=\chi}^{\ell} \frac{\lambda_r}{v_r} \right) \prod_{m=\ell}^{i} e^{-\lambda x/z_{m}} h(t-z/v_m) \prod_{r=\chi}^{\ell} \Gamma_{rm} \prod_{r\neq m}^{i} \Gamma_{rm} \times \sum_{r=\ell}^{i} \sum_{j=1}^{\ell} \frac{B_{j1} B_{\ell j}}{\Gamma_{rm}} \left\{ e^{-\Delta_{rm}(t-z/v_m)} - e^{-\lambda x/z_{m}} \right\} \right]
\]

With the help of the illustration at the end of section 4.4.2 for three members, the above solution is as follows:

\[
N_i^0(z,t) = e^{-\lambda x/z_{v_i}} h(t-z/v_1) \sum_{j=1}^{i-1} B_{ij} \sum_{m=\ell}^{i} e^{-\lambda x/z_{m}} h(t-z/v_m) \]

\[
N_i^0(z,t) = e^{-\lambda x/z_{v_i}} h(t-z/v_1) \left[ \begin{array}{c}
\lambda_1 \\
v_1
\end{array} \right] \]

\[
N_i^0(z,t) = e^{-\lambda x/z_{v_i}} h(t-z/v_1) \left[ \begin{array}{c}
\lambda_1 \\
v_1
\end{array} \right] \frac{B_{21} e^{-\lambda x/z_{v_1}}}{\Gamma_{21}} \left[ \begin{array}{c}
\Delta_{21} \lambda_2 \\
\Delta_{21} v_1
\end{array} \right] \frac{B_{11} e^{-\lambda x/z_{v_1}}}{\lambda_1 - \lambda_{12}} \frac{B_{11} e^{-\lambda x/z_{v_1}}}{\lambda_1 - \lambda_{12}} \left[ \begin{array}{c}
\Delta_{12} \lambda_2 \\
\Delta_{12} v_1
\end{array} \right] \left[ \begin{array}{c}
\Delta_{12} \lambda_2 \\
\Delta_{12} v_1
\end{array} \right] \right]
\]

(5.4)
\[ N^p_3(\tau_z, t) = e^{-\lambda_3 z/v_3} h(t-z/v_3) \left[ B_{31} e^{-\lambda_1 (t-z/v_3)} + B_{32} e^{-\lambda_2 (t-z/v_3)} + B_{33} e^{-\lambda_3 (t-z/v_3)} \right] \]

\[
\begin{align*}
+ \frac{1}{v_1 v_2} h(t-z/v_1) & \quad \frac{e^{-\lambda_1 z/v_1}}{\Gamma_3} \left[ B_{31}^\prime \frac{B_{11}^\prime}{B_{11}^\prime - \Delta_{21}} \begin{bmatrix} \frac{D_{13} B_{11}}{1 - \Delta_{13}} & e^{-\lambda_2 (t-z/v_2)} & -e^{-\lambda_1 (t-z/v_1)} \\ e^{-\lambda_2 (t-z/v_2)} & e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_2 (t-z/v_2)} \\ -e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_2 (t-z/v_2)} & -e^{-\lambda_1 (t-z/v_1)} \end{bmatrix} \right] \\
+ \frac{1}{v_1 v_2} h(t-z/v_2) & \quad \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{12} \Gamma_{32}} \left[ B_{12}^\prime \frac{B_{11}^\prime}{B_{11}^\prime - \Delta_{12}} \begin{bmatrix} \frac{D_{23} B_{11}}{1 - \Delta_{23}} & e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \\ e^{-\lambda_3 (t-z/v_3)} & e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} \\ -e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \end{bmatrix} \right] \\
+ \frac{1}{v_1 v_2} h(t-z/v_3) & \quad \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{13} \Gamma_{23}} \left[ B_{13}^\prime \frac{B_{11}^\prime}{B_{11}^\prime - \Delta_{13}} \begin{bmatrix} \frac{D_{32} B_{11}}{1 - \Delta_{32}} & e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \\ e^{-\lambda_3 (t-z/v_3)} & e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} \\ -e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \end{bmatrix} \right] \\
+ \frac{1}{v_2} h(t-z/v_2) & \quad \frac{e^{-\lambda_2 z/v_2}}{\Gamma_{32}} \left[ B_{32}^\prime \frac{B_{31}^\prime}{B_{31}^\prime - \Delta_{32}} \begin{bmatrix} \frac{D_{23} B_{31}}{1 - \Delta_{23}} & e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \\ e^{-\lambda_3 (t-z/v_3)} & e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} \\ -e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \end{bmatrix} \right] \\
+ \frac{1}{v_2} h(t-z/v_3) & \quad \frac{e^{-\lambda_3 z/v_3}}{\Gamma_{23}} \left[ B_{23}^\prime \frac{B_{21}^\prime}{B_{21}^\prime - \Delta_{23}} \begin{bmatrix} \frac{D_{32} B_{21}}{1 - \Delta_{32}} & e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \\ e^{-\lambda_3 (t-z/v_3)} & e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} \\ -e^{-\lambda_1 (t-z/v_1)} & -e^{-\lambda_3 (t-z/v_3)} & -e^{-\lambda_1 (t-z/v_1)} \end{bmatrix} \right] \\
\end{align*}
\]

(5.5)
where

\[ B_{11} = N_1^0, \quad B_{21} = \frac{K_1 \lambda_1 N_1^0}{K_1 (\lambda_1 - \lambda_1)}, \quad B_{22} = N_2^0 - \frac{K_2 \lambda_2 N_2^0}{K_1 (\lambda_1 - \lambda_1)}, \quad B_{31} = \frac{K_3 \lambda_3 N_1^0}{K_1 (\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \]

\[ B_{32} = \frac{K_3 \lambda_3 N_2^0}{K_1 (\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} + \frac{K_3 \lambda_3 N_1^0}{K_2 (\lambda_3 - \lambda_2)} \]

\[ B_{33} = \frac{K_3 \lambda_3 N_3^0}{K_2 (\lambda_3 - \lambda_2)} + \frac{K_3 \lambda_3 N_2^0}{K_2 (\lambda_3 - \lambda_2)} + n_3^0 \]

(5.6)

\[ \Lambda_1 = \lambda_1 + \lambda_1^*; \quad \Gamma_{rm} = -\Gamma_{mr} = \frac{v_m - v_r}{v_r - v_m}; \quad \delta_{rm} = \Delta_{mr} = \frac{\lambda_{r,m} - \lambda_{r,m}^*}{v_m - v_r} \]

\[ D_{31}^{\text{31}} = -\frac{1}{\delta_{31}^{\text{31}} - \delta_{21}^{\text{21}}}; \quad D_{12}^{\text{31}} = -\frac{1}{\delta_{32}^{\text{32}} - \delta_{12}^{\text{12}}} \]

\[ D_{13}^{\text{31}} = -\frac{1}{\delta_{23}^{\text{23}} - \delta_{13}^{\text{13}}}; \quad D_{32}^{\text{32}} = D_{23}^{\text{32}} = 1; \quad D_{21}^{\text{21}} = D_{12}^{\text{12}} = 1 \]

5.2.2 Solution for exponential release

The solution for the exponential release mode is expressed by setting \( k_i = k \) in (3.23).

\[ N_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_i t} \cdot h(t) \]

\[ B_{ij} = \sum_{m=1}^{j} N_m^0 \left[ \prod_{r=m}^{i} \frac{\lambda_r}{\lambda_i} \right] \left[ \prod_{k=m}^{i} \frac{(\lambda_k - \lambda_i)}{(\lambda_k - \lambda_j)} \right] \]

(5.7)
The concentration of the $i^{th}$ nuclide can then be obtained by setting $K_i = k$ in (5.2) and replacing $B_{ij}'$ by $B_{ij}$:

$$N_i^e(z, t) = e^{-\lambda_1 z/v_1} h(t-z/v_1) \sum_{j=1}^{i} B_{ij} e^{-(\lambda_j + k)(t-z/v_1)}$$

$$+ \sum_{l=1}^{i-1} \left( \prod_{r=l}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=l}^{i} \frac{e^{-\lambda_m z/v_m} h(t-z/v_m)}{\prod_{r=l}^{i} \frac{\Gamma_{rm}}{\Gamma_{rm}}} \sum_{j=1}^{i} B_{lj} e^{-(\lambda_j + k)(t-z/v_m)}$$

$$\times \sum_{j=1}^{i} \frac{D_{rl} B_{lj}}{B_{ij}} \left\{ \frac{-\Delta_{rm}(t-z/v_m)}{\lambda_j + k - \Delta_{rm}} + e^{-(\lambda_j + k)(t-z/v_m)} \right\}$$

(5.8)

For three members, we have

$$N_1^e(z, t) = B_{11} h(t-z/v_1) e^{-(\lambda_1 + k)t + kz/v_1}$$

(5.9)

$$N_2^e(z, t) = e^{-\lambda_2 z/v_2} h(t-z/v_2) \left[ B_{21} e^{-(\lambda_1 + k_1)(t-z/v_1)} + B_{22} e^{-(\lambda_1 + k_2)(t-z/v_2)} \right]$$

$$+ \frac{\lambda_1}{v_1} h(t-z/v_1) e^{\frac{-\lambda_1 z/v_1}{\Gamma_{21}}} \frac{D_{21} B_{11}}{(k+\lambda_1 - \Delta_{21})}$$

$$\times \left[ e^{-\Delta_{21}(t-z/v_1)} - (\lambda_1 + k)(t-z/v_1) \right]$$

$$+ \frac{\lambda_1}{v_1} h(t-z/v_2) e^{\frac{-\lambda_1 z/v_2}{\Gamma_{12}}} \frac{D_{12} B_{11}}{(k+\lambda_1 - \Delta_{12})}$$

$$\times \left[ e^{-\Delta_{12}(t-z/v_2)} - (\lambda_1 + k)(t-z/v_2) \right]$$

(5.10)
\begin{align*}
\eta_3^{(z,t)}(z,t) &= e^{-\lambda_3 z/v_3} \eta_t(z/v_3) \left[ B_{311} e^{-(\lambda_1 + k)(t-z/v_3)} + B_{322} e^{-(\lambda_2 + k)(t-z/v_3)} + B_{333} e^{-(\lambda_3 + k)(t-z/v_3)} \right] \\
&\quad + \frac{\lambda_1 \lambda_2}{v_1 v_2} h_t(z/v_1) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{21}} \right] e^{-\lambda_1 z/v_1} \left[ e^{-\Delta_{21}(t-z/v_1)} - e^{-(\lambda_1 + k)(t-z/v_1)} \right] \\
&\quad + \frac{\lambda_1 \lambda_2}{v_1 v_2} h_t(z/v_2) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{31}} \right] e^{-\lambda_1 z/v_2} \left[ e^{-\Delta_{31}(t-z/v_1)} - e^{-(\lambda_1 + k)(t-z/v_1)} \right] \\
&\quad + \frac{\lambda_2 \lambda_2}{v_1 v_2} h_t(z/v_3) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{32}} \right] e^{-\lambda_1 z/v_3} \left[ e^{-\Delta_{32}(t-z/v_2)} - e^{-(\lambda_1 + k)(t-z/v_2)} \right] \\
&\quad + \frac{\lambda_1 \lambda_2}{v_1 v_2} h_t(z/v_3) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{33}} \right] e^{-\lambda_1 z/v_3} \left[ e^{-\Delta_{33}(t-z/v_3)} - e^{-(\lambda_1 + k)(t-z/v_3)} \right] \\
&\quad + \frac{\lambda_2 \lambda_2}{v_2} h_t(z/v_2) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{23}} \right] e^{-\lambda_2 z/v_2} \left[ e^{-\Delta_{23}(t-z/v_2)} - e^{-(\lambda_2 + k)(t-z/v_2)} \right] \\
&\quad + \frac{\lambda_2 \lambda_2}{v_2} h_t(z/v_3) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{23}} \right] e^{-\lambda_2 z/v_3} \left[ e^{-\Delta_{23}(t-z/v_3)} - e^{-(\lambda_2 + k)(t-z/v_3)} \right] \\
&\quad + \frac{\lambda_2 \lambda_2}{v_2} h_t(z/v_3) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{23}} \right] e^{-\lambda_2 z/v_3} \left[ e^{-\Delta_{23}(t-z/v_3)} - e^{-(\lambda_2 + k)(t-z/v_3)} \right] \\
&\quad + \frac{\lambda_2 \lambda_2}{v_2} h_t(z/v_3) \left[ \frac{D_{311} B_{111}}{\lambda_1 + k - \Delta_{23}} \right] e^{-\lambda_2 z/v_3} \left[ e^{-\Delta_{23}(t-z/v_3)} - e^{-(\lambda_2 + k)(t-z/v_3)} \right]
\end{align*}

(5.11a)
Where

\[ B_{11} = N_1^0, \quad B_{21} = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1}, \quad B_{22} = N_2^0 - \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} \]

\[ B_{31} = \frac{\lambda_1 \lambda_2 N_1^0}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}, \quad B_{32} = \frac{\lambda_1 \lambda_2 N_1^0}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{\lambda_2 N_2^0}{(\lambda_3 - \lambda_2)} \]

\[ B_{33} = \frac{\lambda_1 \lambda_2 N_2^0}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} + \frac{\lambda_2 N_2^0}{\lambda_2 - \lambda_3} + N_3^0 \quad (5.11b) \]

### 5.2.3 Solution for step release

The solution for the step release mode is easily obtained from the solution of exponential release mode by setting \( k = 0 \).

\[ N_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} h(t) \quad (5.12) \]

From (5.8) the concentration for the \( i \)-th nuclide is given by:

\[ N_i^s(z,t) = e^{-\lambda_1 z/v_1} h(t-z/v_1) \sum_{j=1}^{i} B_{ij} e^{-\lambda_j (t-z/v_1)} \]

\[ + \sum_{\ell=1}^{i-1} \left( \prod_{r=\ell}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=\ell}^{i} \frac{e^{-\lambda_m z/v_m}}{\prod_{r=\ell}^{i} \Gamma_{rm}} h(t-z/v_m) \]

\[ \times \sum_{r=\ell}^{i} \sum_{j=1}^{\ell} \frac{D_{rm} p_{lj}}{\lambda_j - \Delta_{rm}} \left\{ e^{-\Delta_{rm} (t-z/v_m)} - e^{-\lambda_j (t-z/v_m)} \right\} \]

\[ \quad + \sum_{\ell=1}^{i-1} \left( \prod_{r=\ell}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=\ell}^{i} \frac{e^{-\lambda_m z/v_m}}{\prod_{r=\ell}^{i} \Gamma_{rm}} h(t-z/v_m) \]

\[ \times \sum_{r=\ell}^{i} \sum_{j=1}^{\ell} \frac{D_{rm} p_{lj}}{\lambda_j - \Delta_{rm}} \left\{ e^{-\Delta_{rm} (t-z/v_m)} - e^{-\lambda_j (t-z/v_m)} \right\} \]
For three members, we have:

\[ N_1^S(z,t) = N_1^0 e^{-\lambda_1 t} h(t-z/v_1) \]  \hspace{1cm} (5.14)

\[ N_2^S(z,t) = e^{-\lambda_2 z/v_2} h(t-z/v_2) \left[ B_{21} e^{-\lambda_1(t-z/v_2)} + B_{22} e^{-\lambda_2(t-z/v_2)} \right] \]

\[ + \frac{\lambda_1}{v_1} h(t-z/v_1) e^{-\lambda_1 z/v_1} \frac{B_{21} B_{11}}{\Gamma_{21} (\lambda_1 - \lambda_{21})} \left[ e^{-\Delta_{21}(t-z/v_1)} - e^{-\lambda_1(t-z/v_1)} \right] \]

\[ + \frac{\lambda_1}{v_1} h(t-z/v_2) e^{-\lambda_2 z/v_2} \frac{B_{21} B_{11}}{\Gamma_{12} (\lambda_1 - \lambda_{12})} \left[ e^{-\Delta_{21}(t-z/v_2)} - e^{-\lambda_1(t-z/v_2)} \right] \]

(5.15)
\[ n_3^b(z,t) = e^{-\lambda_3 z / v_3} h(t-z / v_3) \left[ b_{31} e^{-\lambda_1 (t-z / v_3)} + b_{32} e^{-\lambda_2 (t-z / v_3)} + b_{33} e^{-\lambda_3 (t-z / v_3)} \right] \]

\[ + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t-z / v_1) \frac{e^{-\lambda_1 z / v_1}}{t_{21}^{\lambda_1}} \begin{cases} \beta_{31} & \begin{cases} e^{-\Delta_{12} (t-z / v_2)} - e^{-\lambda_1 (t-z / v_1)} \\ e^{-\Delta_{12} (t-z / v_2)} - e^{-\lambda_1 (t-z / v_1)} \end{cases} \\ \lambda_1 - \lambda_2 \end{cases} \]

\[ + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t-z / v_2) \frac{e^{-\lambda_2 z / v_2}}{t_{12}^{\lambda_2}} \begin{cases} \beta_{32} & \begin{cases} e^{-\Delta_{12} (t-z / v_2)} - e^{-\lambda_1 (t-z / v_2)} \\ e^{-\Delta_{23} (t-z / v_2)} - e^{-\lambda_2 (t-z / v_2)} \end{cases} \\ \lambda_1 - \lambda_2 \end{cases} \]

\[ + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t-z / v_3) \frac{e^{-\lambda_3 z / v_3}}{t_{13}^{\lambda_2}} \begin{cases} \beta_{33} & \begin{cases} e^{-\Delta_{13} (t-z / v_3)} - e^{-\lambda_1 (t-z / v_3)} \\ e^{-\Delta_{23} (t-z / v_3)} - e^{-\lambda_2 (t-z / v_3)} \end{cases} \\ \lambda_1 - \lambda_2 \end{cases} \]

\[ + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t-z / v_2) \frac{e^{-\lambda_2 z / v_2}}{t_{32}^{\lambda_2}} \begin{cases} \beta_{32} & \begin{cases} e^{-\Delta_{32} (t-z / v_2)} - e^{-\lambda_2 (t-z / v_2)} \\ e^{-\Delta_{32} (t-z / v_2)} - e^{-\lambda_2 (t-z / v_2)} \end{cases} \\ \lambda_2 - \Delta_{32} \end{cases} \]

\[ + \frac{\lambda_1 \lambda_2}{v_1 v_2} h(t-z / v_3) \frac{e^{-\lambda_3 z / v_3}}{t_{23}^{\lambda_2}} \begin{cases} \beta_{33} & \begin{cases} e^{-\Delta_{32} (t-z / v_3)} - e^{-\lambda_2 (t-z / v_3)} \\ e^{-\Delta_{32} (t-z / v_3)} - e^{-\lambda_2 (t-z / v_3)} \end{cases} \\ \lambda_2 - \Delta_{32} \end{cases} \]

(5.16)

5.2.4 Solution for band release

The band release mode is defined by

\[ N_1(t) = b_1(t)[h(t) - h(t-T)] \]  

(5.17)
The solution for this release mode can be obtained from the solution
of step release mode by using the superposition relation given by
(3.60).

\[ N^b_i(t) = N^S(z,t; B_{ij}) - N^S(z,t-T; B_{ij} e^{-\lambda_j T}) \]  

(3.60)

The first term in the right hand side is given by (5.13). The second
term can be obtained by replacing \( B_{ij} \) in (5.13) by \( B_{ij} \times e^{-\lambda_j T} \) and
also replacing \( t \) by \( t-T \). The resultant solution is expressed as:

\[ N^b_i(z,t) = e^{-\lambda_i z/v_i} \sum_{j=1}^{i} B_{ij} e^{-\lambda_j (t-z/v_i)} \left\{ h(t-z/v_i) - h(t-T-z/v_i) \right\} \]

\[ + \sum_{k=1}^{i-1} \left( \prod_{r=k}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=k}^{i} e^{-\lambda_m z/v_m} \sum_{r=k}^{i} \frac{1}{\lambda_j - \Delta_{rm}} \sum_{j=1}^{l} D_{rm} B_{kj} \]

(5.18)

\[ \times \left[ e^{-\Delta_{rm} (t-z/v_m)} \left\{ h(t-z/v_m) - e^{-(\lambda_j - \Delta_{rm}) T} h(t-T-z/v_m) \right\} \right. \]

\[ - e^{-\lambda_j (t-z/v_m)} \left\{ h(t-z/v_m) - h(t-T-z/v_m) \right\} \]

For three members, the solution can be obtained with the help of (5.14),
(5.15), and (5.16).
5.2.5 Solution for impulse release

The impulse release mode is a limiting case which can be obtained by applying the condition, \( \lim_{T \to 0} \frac{1}{T} [h(t-z/v_1) - h(t-T-z/v_1)] = \delta(t-z/v_1) \) on the solution of band release mode. Then, the concentration of the \( i \)th nuclide is expressed as:

\[
N_i^I(z, t) = (N_1^0 T)e^{-\frac{\lambda_1 z}{v_1}} \delta(t-z/v_1)
\]

\[
+ \sum_{\ell=1}^{i-1} \left( \prod_{r=\ell}^{i-1} \frac{\lambda_r}{v_r} \right) \sum_{m=\ell}^{i} \frac{N_{i}^{0} \lambda_{m} z/v_{m}}{\prod_{r=\ell}^{i} \Gamma_{rm}} \frac{h(t-z/v_{m})}{h(t)}
\]

\[
\times \sum_{r=\ell}^{i} D_{i\ell}^{r\ell} e^{-\Delta_{rm}(t-z/v_{m})}
\]

With the help of the illustration at the end of section 4.4.2 for three members, we have:

\[
N_1^I(z, t) = (N_1^0 T)e^{-\frac{\lambda_1 z}{v_1}} \delta(t-z/v_1)
\]
\[ N_2^T(z, t) = (N_2^T e^{\frac{-\lambda_2 z}{v_2}}) \delta(t-z/v_2) \]

\[ \frac{\lambda_1}{v_1} \frac{N_1^T}{\Gamma_{21}} e^{\frac{-\lambda_1 z}{v_1}} h(t-z/v_1) \Delta_{21}(t-z/v_1) \]

\[ \frac{\lambda_1}{v_1} \frac{N_1^T}{\Gamma_{12}} e^{\frac{-\lambda_2 z}{v_2}} h(t-z/v_2) \Delta_{12}(t-z/v_2) \]

\[ N_3^T(z, t) = (N_3^T e^{\frac{-3z}{v_3}}) \delta(t-z/v_3) \]

\[ \frac{\lambda_1 \lambda_2}{v_1 v_2} \frac{N_1 e^{\frac{-\lambda_1 z}{v_1}}}{\Gamma_{21} \Gamma_{23}} h(t-z/v_1) \left\{ D_{12}^1 e^{-\Delta_{12}(t-z/v_2)} + D_{31}^1 e^{-\Delta_{31}(t-z/v_1)} \right\} \]

\[ \frac{\lambda_1 \lambda_2}{v_1 v_2} \frac{N_1 e^{\frac{-\lambda_2 z}{v_2}}}{\Gamma_{12} \Gamma_{32}} h(t-z/v_2) \left\{ D_{12}^3 e^{-\Delta_{12}(t-z/v_2)} + D_{32}^3 e^{-\Delta_{32}(t-z/v_2)} \right\} \]

\[ \frac{\lambda_1 \lambda_2}{v_1 v_2} \frac{N_1 e^{\frac{-\lambda_2 z}{v_2}}}{\Gamma_{13} \Gamma_{23}} h(t-z/v_3) \left\{ D_{13}^1 e^{-\Delta_{13}(t-z/v_3)} + D_{23}^1 e^{-\Delta_{23}(t-z/v_3)} \right\} \]

5.3 Application of the General Solution—Solution of the Transport Equation for a Three-Member Decay Chain with Dispersion, Using the Plane-Source Boundary Condition (H1)

The general solution for the transport equation with plane source and dispersion is given by (4.38).
\[ N_2(z, t) = \int_0^t f_2(t - \theta_2) e^{-\lambda_2 \theta_2} F(v_2 \theta_2, z - v_2 \theta_2) d\theta_2 \]

\[ + \frac{\lambda_1 v_2}{v_1} \int_0^t \int_0^t d\theta_2 f_1(t - \theta_1) e^{-\lambda_1 \theta_1 - \lambda_2 \theta_2} \]

\[ \times F(v_1 \theta_1 + v_2 \theta_2, z - v_1 \theta_1 - v_2 \theta_2) \]
\[ n_3(z, t) = \int_0^t f_3(t-\theta_3)e^{-\lambda_3 \theta_3} F(v_3 \theta_3, z-v_3 \theta_3) d\theta_3 \]
\[ + \frac{\lambda_2 \nu_3}{\nu_2} \int_0^t d\theta_3 \int_0^{t-\theta_3} d\theta_2 f_2(t-\theta_2-\theta_3)e^{-\lambda_2 \theta_2-\lambda_3 \theta_2} F(v_2 \theta_2+v_3 \theta_3, z-v_2 \theta_2-v_3 \theta_3) \]
\[ = \frac{\lambda_1 \lambda_2 \nu_3}{\nu_1} \int_0^t d\theta_3 \int_0^{t-\theta_3} d\theta_2 \int_0^{t-\theta_2-\theta_3} d\theta_1 f_1(t-\theta_1-\theta_2-\theta_3)e^{-\lambda_1 \theta_1-\lambda_2 \theta_2-\lambda_3 \theta_2} \]
\[ \times F(v_1 \theta_1+v_2 \theta_2+v_3 \theta_3, z-v_1 \theta_1-v_2 \theta_2-v_3 \theta_3) \]

(5.25)

where \( f_i(t) \) is the source term at the repository. It can be related to the concentration at the repository with the use of (3.33) and (4.23):

\[ f_1(t) = N_1(t) \nu_1 \]

(5.26)

In this section we apply (5.23) - (5.25) to a three-member chain and propose explicit solutions for step and band release modes.

In the decaying step-release mode \( N_1(t) \) is expressed by the Bateman equation. In this case, \( f_1(t) \) for three members can be obtained as:
\[ f_1(t) = v_1^0 \lambda_1^{-\lambda_1 t} = v_1^0 \lambda_1^{-\lambda_1 t} \]

\[ f_2(t) = v_2 N_2(t) = v_2 \left[ \left( \frac{N_2^0}{\lambda_1} + \frac{N_1^0}{\lambda_1 - \lambda_2} \right) e^{-\lambda_2 t} + \frac{N_1^0}{\lambda_1 - \lambda_2} e^{-\lambda_1 t} \right] \]

\[ \leq v_2 \left[ \frac{b_{22} e^{-\lambda_2 t} + b_{21} e^{-\lambda_1 t}}{} \right] \]

\[ f_3(t) = v_3 N_3(t) = v_3 \left[ \left( \frac{N_3^0}{\lambda_2} + \frac{N_1^0 \lambda_2}{\lambda_2 - \lambda_3} + \frac{N_1^0 \lambda_1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \right) e^{-\lambda_3 t} \right. \]

\[ + \left. \left( + \frac{N_2^0 \lambda_2}{\lambda_3 - \lambda_2} + \frac{N_1^0 \lambda_1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \right) e^{-\lambda_2 t} \right. \]

\[ + \left. \frac{N_1^0 \lambda_1}{(\lambda_1 - \lambda_3)(\lambda_1 - \lambda_2)} e^{-\lambda_1 t} \right] \]

\[ \leq v_3 \left[ \frac{b_{33} e^{-\lambda_3 t} + b_{32} e^{-\lambda_2 t} + b_{31} e^{-\lambda_1 t}}{} \right] \]

(5.27)

The concentrations for three members can be obtained by introducing (5.27) into (5.23) - (5.25), integrating by parts, and using the following relation:

\[ \frac{2}{\sqrt{\pi}} \int_0^{a/b} e^{-\left( \lambda^2 + a^2/\lambda^2 \right)} d\lambda = \frac{1}{2} \left[ e^{-2a} \text{erfc}(b-a/b) - e^{2a} \text{erfc}(b+a/b) \right] \] (5.28)

The resultant expressions for the concentrations are:

\[ N_1^S(z, t) = B_{11} E(1,1;1) \] (5.29)
\[ N_2^S(z, t) = B_{22} E(2, 2; 2) + B_{21} E(1, 1; 2) \]
\[ + \frac{B_{11} \lambda_1}{v_1 \Delta_{12}^{-1} \lambda_1 \Gamma_{12}} \left[ E(1, 1; 2) - E(1, 1; 1) \right] \]
\[ \Lambda_{kj} = \frac{\lambda_k}{v_k} - \frac{\lambda_j}{v_j}, \quad \Gamma_{kj} = \frac{1}{v_k} - \frac{1}{v_j} \]

\[
\beta_{ij} = \begin{cases} 
\lambda_i, & i=j \\
\Lambda_{ij}/\Gamma_{ij}, & i\neq j
\end{cases}
\] (5.32)

\[ \gamma_{ijk} = 1 + 4\kappa(\lambda_k - \beta_{ij})/v_k \]

\[
E(i,j;k) = e^{\beta_{ij} t + z/2\kappa} \int_0^{\sqrt{v_k}t/4\kappa} \frac{2\sqrt{\gamma_{ijk}}}{\sqrt{\pi}} \exp \left[ -\left( \frac{y^2}{\gamma_{ijk}} \right) \right] dy
\]

\[ = e^{\beta_{ij} t + z/2\kappa} \left[ \frac{1}{2\gamma_{ijk}} e^{-\frac{|z|}{2\kappa}\sqrt{\gamma_{ijk}}} \text{erfc} \left( \frac{|z| - \frac{v_k t}{\sqrt{\gamma_{ijk}}}}{\sqrt{4\kappa v_k t}} \right) \right] \] (5.33)

\[ \gamma_{ijk} > 0 \]

In compact form, the above equations can be described as:

\[ n_1(z,t) = B_{11} E(1,1;1) \]

\[ n_2(z,t) = \sum_{j=1}^{2} B_{2j} E(j,j;2) + \sum_{j=1}^{2} B_{11} \lambda_j \frac{\Lambda_{12}}{v_1^2} \frac{\Lambda_{21}}{v_2^2} \left[ E(1,1;2) - E(1,1;1) + E(2,2;1) - E(1,2;2) \right] \]

\[ n_3(z,t) = \sum_{j=1}^{3} B_{3j} E(j,j;3) + \frac{\lambda_2}{v_2} \sum_{j=1}^{2} \frac{B_{2j}}{v_1^2} \left[ E(j,j;3) - E(j,j;2) + E(2,3;2) - E(2,3;3) \right] \]

\[ + \frac{\lambda_1 \lambda_2 B_{11}}{v_1^2} \sum_{j=1}^{3} \left[ \frac{E(1,1;j)}{(\Lambda_{1j} - \lambda_j \frac{\Gamma_{1j}}{v_1}) (\Lambda_{j1} - \lambda_j \frac{\Gamma_{j1}}{v_1})} + \frac{\Gamma_{kj} E(k,j;j)}{(\Lambda_{k1} - \lambda_j \frac{\Gamma_{kj}}{v_1}) (\Lambda_{jk} - \lambda_j \frac{\Gamma_{kj}}{v_1})} \right] \]

\[ + \frac{\Gamma_{kj} E(k,j;j)}{(\Lambda_{k1} - \lambda_j \frac{\Gamma_{kj}}{v_1}) (\Lambda_{jk} - \lambda_j \frac{\Gamma_{kj}}{v_1})} \]

\[ (k < 2, \quad k \neq j, \quad j \neq k; \quad k = 1, 2, 3) \] (5.34)
To obtain the band-release solutions, we apply the superposition relation described by (3.60):

\[ N_i^b(z,t) = N_i^a(z,t; B_{ij}) - N_i^s(z,t-T; B_{ij}^{-\lambda_j T}) \]  

(5.35)

The first term on the right-hand side is given by (5.34). The second term is obtained by replacing \( B_{ij} \) in (5.34) by \( B_{ij} \exp(-\lambda_j T) \) and also replacing \( t \) by \( (t-T) \). The concentrations for a band release can be obtained from (5.34) and (5.35).

When \( \kappa \to 0 \), (5.34) agrees with the solutions for no dispersion. Thus (5.34) includes the case of no dispersion.

5.4 Application of the General Solution--Solution of the Transport Equation for a Three-Member Decay Chain with Dispersion, Using the Concentration Boundary Condition (F2)

The governing equation of one-dimensional transport, with local chemical equilibrium is

\[ \eta_1 \frac{\partial^2 N_1}{\partial z^2} - v_1 \frac{\partial N_1}{\partial z} - \frac{\partial N_1}{\partial t} - \lambda_1 N_1 = -\lambda_{i-1} N_{i-1} - \frac{K_{i-1} N_{i-1}}{K_1} \]  

(4.2)

\[ \lambda_i = 0, \quad i = 1, 2, ... \]

The initial and boundary conditions are

\[ N_i(z,0) = 0, \quad N_i(\infty, t) = \text{Order of } e^{-kz}, \quad N_i(0,t) = N_i^0 \phi_i(t) \]  

(5.36)
The recursive solution of (4.2) and (5.36) is given in the preceding chapter by (4.61), which can be rewritten as

\[ N_i(z,t) = \exp \left[ \frac{z}{2\kappa} - \left( \lambda_i + \frac{v_i}{4\kappa} \right) t \right] \]

\[ \times \int_0^\infty -\kappa v_i p^2 t \sqrt{\frac{2}{\pi}} \sin(pz) \int_0^t e^{\frac{\kappa v_i p^2}{2} \theta} R_{i-1}(p,\theta) d\theta dp \]  

(5.37)

where

\[ R_{i-1}(p,\theta) = \kappa v_i S_{i-1}(p,t) + v_i \kappa \cos(pz) \exp \left\{ \left( \lambda_i + \frac{v_i}{4\kappa} \right) t \right\} \times \kappa^0 \theta_1(\theta) \]

\[ S_{i-1}(p,t) = \int_0^\infty \sqrt{\frac{2}{\pi}} \sin(pz) e^{-\beta_i t - \frac{z}{2\kappa}} N_{i-1}(z,t) \frac{\lambda_i - \frac{1}{v_i}}{v_i} dz \]  

(5.38)

Here we are concerned with the concentrations of a three-member decay chain for the step-release and band-release modes.

For a step release, the concentration boundary condition at the repository is expressed as
$$N_i(0,t) = N_i^0 \phi_i(t) = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t} h(t)$$

(5.39)

The concentrations of the three members can then be calculated from (5.37) – (5.39) by an iterative procedure, with the help of a table of Fourier integrals. The resultant concentrations are reduced to the analytical solution (5.34) for a step release in the preceding section, but with the function $E(i,j;k)$ defined as

$$E(i,j;k) = e^{z/2\kappa - \beta_{1j} t} \int_{\beta_{1j} t}^{\infty} \sqrt{\pi} \exp \left[ - \left( y^2 + \frac{z^2 \gamma_{1jk}}{16\kappa^2 y^2} \right) \right] dy$$

$$= \frac{1}{2} e^{z/2\kappa - \beta_{1j} t} \left[ z \sqrt{\gamma_{1jk}} / 2\kappa \right] \text{erfc} \left[ \frac{z + \sqrt{\gamma_{1jk}} / 2\kappa}{\sqrt{4\kappa v_k t}} \right]$$

$$+ e^{-z \sqrt{\gamma_{1jk}} / 2\kappa} \text{erfc} \left[ \frac{z - \sqrt{\gamma_{1jk}} / 2\kappa}{\sqrt{4\kappa v_k t}} \right] \quad \{(\gamma_{1jk} > 0)\}

(5.40)

Given this step-release solution, the band-release solution can be obtained by applying the superposition Equation (5.35).

When dispersion is neglected the functions $E(k,j;k)$ i.e., (5.40) and (5.33) agree with each other. Thus, the solutions for the concentration boundary condition agree with the solutions for the plane-source boundary condition. This would be expected on physical grounds. In the absence
of dispersion, the plane-source boundary condition yields directly the concentration boundary condition. When dispersion is finite, the plane-source boundary condition is the proper formulation. The concentration boundary condition remains an approximation for no dispersion at the point of dissolution of the waste.

5.5 Migration Behavior of the Decay Chain $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ (H4)

One of the important nuclide chains to be considered in the predictive modeling of hydrogeological transport of radionuclides in high-level wastes is:

$$^{242}\text{Am} \rightarrow ^{242}\text{Cm} \rightarrow ^{238}\text{Pu} \rightarrow ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra} \rightarrow$$

The most important species is usually $^{226}\text{Ra}$, because of its relatively high biological hazard and its relatively high mobility in geologic media. The important time scales for the appearance of $^{226}\text{Ra}$ are of the order of tens of thousands of years, as controlled by the long life of its precursor $^{230}\text{Th}$. Therefore, because of the relatively short lives of the first three members of the above chain, the analysis of $^{226}\text{Ra}$ transport can be reduced to the analysis of the last three members of the above chain, with the assumption that the first three members have already decayed to form the long-lived $^{234}\text{U}$.

In this section, the migration features for this three member decay chain will be elucidated with the help of the solutions of transport equations (5.29) through (5.40) which correspond to the step-release and band-release modes. The computer code is shown in the Appendix.
The three nuclides are denoted by

\[ i = 234\text{U} \]
\[ 2 = 230\text{Th} \]
\[ 3 = 226\text{Ra} \]

5.5.1 Input data and parameters

5.5.1.1 Release mode

In Chapter 3, several release modes have been discussed. In this calculation, we select the band release mode as a typical release mode. The amount of the \( i^{th} \) nuclide \( M_i(t') \) (atoms) in the repository changes with time according to the Bateman equation:

\[
M_i(t') = \sum_{j=1}^{i} B_{ij} e^{-\lambda_j t'} , \quad B_{ij} = \sum_{m=1}^{j} b_m^0 \frac{1}{\lambda_i} \prod_{k=m}^{i} \frac{1}{\lambda_k \prod_{k=m}^{i} (\lambda_k - \lambda_j)} \quad i = 2, 3, \ldots
\]  

(5.41)

Here \( t' \) is the time measured from the moment of initial emplacement of the nuclides in the repository, and we assumed that there are no daughters (nuclides 2 and 3) in the repository at \( t' = 0 \). The change of the amount of each nuclide is calculated as shown in Figure 5.1. The amounts of \( ^{230}\text{Th} \) and \( ^{226}\text{Ra} \) increase with time first, and pass a maximum at \( t' = 2 \times 10^5 \) year. Finally, the ratios of the amounts of the second and the third members relative to that of the first member approach a steady-state (transient equilibrium):
\[
\lim_{t' \to \infty} \frac{M_2(t')}{M_1(t')} = \frac{\lambda_1}{\lambda_2 - \lambda_1} \tag{5.42}
\]

\[
\lim_{t' \to \infty} \frac{M_3(t')}{M_1(t')} = \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}, \text{ since } \lambda_2, \lambda_3 > \lambda_1
\]

To simplify the calculation, we assume two kinds of initial conditions which specify the waste properties at the beginning of dissolution \((t = 0)\): (1) The release of nuclides from the waste to the groundwater occurs at an early stage. (2) The release occurs after the nuclides have reached transient equilibrium. In the first case, pure \(^{234}\text{U}\) is the only nuclide present in the repository, and no daughters are present at the beginning of dissolution \((t = 0)\), so that:

\[
N_2^0 = N_3^0 = 0 \tag{5.43a}
\]

In the second case the initial concentrations \(N_2^0\) and \(N_3^0\) at the beginning of dissolution \((t = 0)\) are assumed from (5.42) to be

\[
N_2^0 = N_1^0 \frac{\lambda_1}{(\lambda_2 - \lambda_1)}, \quad N_3^0 = N_1^0 \frac{\lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \tag{5.43b}
\]
Figure 5.1. Nuclide concentrations versus time in the repository
(z = 0) for the $^{234}\text{U} + ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain.
(Pure $^{234}\text{U}$ source at $t = 0$. $M_i$ = atoms of $i^{th}$ nuclide,
$M_i^0$ = atoms of $^{234}\text{U}$ at $z = 0$, $t = 0.$)
5.5.1.2 Data

Nuclear data and assumed sorption retardation constants (B3) for the nuclides are shown in Table 5.1.

<table>
<thead>
<tr>
<th></th>
<th>(^{234}\text{U})</th>
<th>(^{230}\text{Th})</th>
<th>(^{226}\text{Ra})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Half-life (yr)</td>
<td>(2.44 \times 10^5)</td>
<td>(7.7 \times 10^4)</td>
<td>(1.6 \times 10^3)</td>
</tr>
<tr>
<td>Decay Constant (\lambda_i) (1/yr)</td>
<td>(2.84 \times 10^{-6})</td>
<td>(9.00 \times 10^{-6})</td>
<td>(4.33 \times 10^{-4})</td>
</tr>
<tr>
<td>Sorption Retardation Constant, (K_i)</td>
<td>(1 \times 10^4)</td>
<td>(5 \times 10^4)</td>
<td>(5 \times 10^2)</td>
</tr>
</tbody>
</table>

The values of the parameters, \(K_i\), \(v\) and \(D\) have essentially defined the nature of the sorption medium through which the nuclides migrate. The speed \(v\) of groundwater is assumed to be 100 m/yr. This is of the same order as the measured value for some fractured rock and deep aquifers (H5). The migration speed \(v_i\) of each nuclide \((v_i = v/K_i)\) is:

\[
\begin{align*}
\text{U; } v_1 &= 1.0 \times 10^{-2} \text{ m/yr} \\
\text{Th; } v_2 &= 2.0 \times 10^{-3} \text{ m/yr} \\
\text{Ra; } v_3 &= 2.0 \times 10^{-1} \text{ m/yr}
\end{align*}
\]

In this decay chain, the migration speed of \(^{234}\text{U}\) is faster than that of \(^{230}\text{Th}\), and that of \(^{226}\text{Ra}\) is much faster than its precursors. Half-lives decrease in the order of \(^{234}\text{U}\), \(^{230}\text{Th}\), and \(^{226}\text{Ra}\).
In this section, the successive development of the nuclide profiles are graphically shown for the case of

\[ \text{leach time (T)} = 3 \times 10^4 \text{ yr} \]

This leach time is much longer than the half life of $^{226}$Ra (1600 yr) but much shorter than the time to reach the maximum $M_3/M_1^0$ in Figure 5.1.

During the period of leach time, each of the nuclides firstly dissolved migrates the distance of

- $^{234}$U; $v_1T = 300$ m
- $^{230}$Th; $v_2T = 60$ m
- $^{226}$Ra; $v_3T = 6000$ m

Each nuclide migrates in its half-life the distance shown in Table 5.2

<table>
<thead>
<tr>
<th>Table 5.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Migration length in each half life (half-life) x (migration speed)</td>
</tr>
<tr>
<td>$^{234}$U : 2,440 m</td>
</tr>
<tr>
<td>$^{230}$Th : 154 m</td>
</tr>
<tr>
<td>$^{226}$Ra : 320 m</td>
</tr>
</tbody>
</table>
In spite of its fast migration speed, $^{226}\text{Ra}$ cannot survive long in the absence of its precursor because of its short half-life. On the other hand, $^{234}\text{U}$ can migrate a relatively long distance without significant decay. From Table 5.2, we may expect that this decay chain can propagate over a few kilometers (when $v = 100$ m/yr) because of the long survival distance of the first (parent) nuclide, $^{234}\text{U}$. More precisely, $^{234}\text{U}$ and its daughters can be expected to propagate over a distance $L$ of the order

$$L = \frac{vT}{1/2,1}$$

The value of the dispersive coefficient depends greatly on the water velocity, porosity of the soil column and the pore radius. The characteristic dispersive length, $D/v$, may vary in several orders of magnitude (H5). In the present calculation, a representative value of $D = 10^{-3}$ m$^2$/yr is selected. A value corresponding to the usual molecular diffusion coefficient, $1 \times 10^{-1}$ m$^2$/yr, is also used as a lower-limit estimate.

The concentration profiles can be expected to change with different values of leach time. However, the series of graphs presented, herein for $T = 3 \times 10^4$ yr most of the important qualitative features are expected to be seen.
5.5.2 Concentration profiles of the Decay Chain

\[ {^{234}\text{U}} \rightarrow {^{230}\text{Th}} \rightarrow {^{226}\text{Ra}} \]

5.5.2.1 Comparison of plane-source and concentration boundary conditions, and effect of dispersion

Figure 5.2 shows the plot of the concentration profiles of each nuclide at \( t = 5 \times 10^4 \) yr in the case of pure \( {^{234}\text{U}} \) source in the repository at \( t = 0 \) for two different solutions of the transport equations, i.e., (1) the solution of the transport equation with a plane source and dispersion (Eqs. (5.34) and (5.35); (2) the solution of the transport equation with the concentration boundary condition and dispersion (Eqs. (5.34), (5.35) and (5.40)). In this figure, the broken lines show the solution for the plane-source boundary condition with \( D = 1 \times 10^3 \text{ m}^2/\text{yr} \); the dash-dotted lines show the solution for the concentration boundary condition, also with \( D = 1 \times 10^3 \text{ m}^2/\text{yr} \); and the solid lines show the solution for either boundary condition with \( D = 1 \times 10^{-1} \text{ m}^2/\text{yr} \).

In the case of the small dispersion coefficient, \( D = 1 \times 10^{-1} \text{ m}^2/\text{yr} \) both solutions for the plane source and the concentration boundary agree with each other. Furthermore, it was found by the preliminary calculation (H2, H3) that the calculated concentration profiles almost coincide with those of the dispersion-free case.

In the case of the larger dispersion coefficient, \( D = 1 \times 10^3 \text{ m}^2/\text{yr} \) the concentration profiles near the repository are much more affected by the different boundary conditions. As shown by the curves in broken lines and dash-dotted lines, the near-field concentrations of each nuclide for the concentration boundary condition are much lower than
Figure 5.2. Comparison of concentration profiles for different boundary conditions for the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain at $t = 5 \times 10^4$ yr. (Pure $^{234}\text{U}$ source at $t = 0$.

$N_i(z,t) = \text{atom/cm}^3$, $N_i^0 = \text{atom/cm}^3$ of U at $z = 0$,

$t = 0$, $v = 100 \text{ m/yr}$, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$,

$K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$.)
Figure 5.3. Concentration profiles of $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain
at $t = 1 \times 10^4$ yr. (Pure $^{234}\text{U}$ source at $t = 0$. $N_i(z,t) = \text{atoms/cm}^3$, $N_i^0 = \text{atoms/cm}^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $\kappa_U = 1 \times 10^6$, $\kappa_{\text{Th}} = 5 \times 10^4$, $\kappa_{\text{Ra}} = 5 \times 10^2$.)}
those for the plane source condition. The solution for the plane source condition takes into account the effect of dispersion in the negative z domain, but the solution for the concentration boundary condition neglects this effect. The solution for the transport equation with the plane source condition represents a more realistic model of the transport phenomenon than the solution with the concentration boundary condition which has been usually used by many other researchers. However, the solutions for the two different boundary conditions at locations far from the repository almost agree with each other. The issue of boundary conditions is important only for near-field calculations.

5.5.2.2 Concentration profiles of nuclides in the case of a pure $^{234}$U source in the waste at $t = 0$

Figures 5.3 through 5.6 show the successive development of concentration profiles of each nuclide for the case that the leaching begins at $t = 0$ with no daughter nuclides present. $N_i(z,t)$, $i = 1,2,3$, is the concentration of the $i^{th}$ nuclide in the water phase at a location $z$ and at time $t$. These concentrations were calculated using the plane source boundary conditions.

$N_1^0$ is the concentration of $^{234}$U at $z = 0$ just after beginning of leaching ($N_2^0 = N_3^0 = 0$). $N_1^0$ should be inversely proportional to the leach time, T. The concentrations of $N_i$ are all relative to $N_1^0$, shown by the ratio $N_i/N_1^0$. In these figures, the solid lines show the concentration profiles of nuclides for the dispersion coefficient $D = 1 \times 10^{-1} \text{ m}^2/\text{yr}$, and the broken line are for $D = 1 \times 10^3 \text{ m}^2/\text{yr}$. 
Figure 5.4. Concentration profiles of $^{234}_{\text{U}} \rightarrow ^{230}_{\text{Th}} \rightarrow ^{226}_{\text{Ra}}$ decay chain at $t = 5 \times 10^4$ yr. (Pure $^{234}_{\text{U}}$ source at $t = 0$. $N_i(z,t) =$ atoms/cm$^3$, $N_i^0 =$ atoms/cm$^3$ of $^{234}_{\text{U}}$ at $z = 0$, $t = 0$, $v =$ 100 m/yr, $T = 3 \times 10^4$ yr, $K_u = 1 \times 10^4$. $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$. )
Figure 5.5. Concentration profiles of $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain at $t = 1 \times 10^5 \text{ yr}$. (Pure $^{234}\text{U}$ source at $t = 0$. $N_i(z,t) = \text{atoms/cm}^3$, $N_o^1 = \text{atoms/cm}^3$ of $^{234}\text{U}$ at $z = 0$, $v = 100 \text{ m/yr}$, $T = 3 \times 10^4 \text{ yr}$, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
Figure 5.6. Concentration profiles of $^{234}$U $\rightarrow$ $^{230}$Th $\rightarrow$ $^{226}$Ra decay chain at $t = 2 \times 10^5$ yr. (Pure $^{234}$U source at $t = 0$, $N_i(z,t)$ = atoms/cm$^3$, $N_0^o$ = atoms/cm$^3$ of $^{234}$U at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
In Figure 5.3, $t$ is less than the leach time, $T = 3 \times 10^4$ yr.

$^{230}$Th which was first formed and leached from the waste has now reached the location $z = v_2 t$. Let us first consider the case of a small dispersion coefficient. All $^{230}$Th in the region $v_2 t < z < v_1 t$ has been produced by the decay of $^{234}$U already leached from the waste, except a very small amount contributed by dispersive transport. In the region $v_2 t < z < v_1 t$, the concentration of $^{230}$Th decreases with distances, and ends at the leading edge of $^{234}$U band, i.e., $z = v_1 t$.

In the region of $z < v_2 t$, there are two contributions to the concentration of $^{230}$Th. One is the decay of $^{234}$U that has been leached out from the repository. The other is $^{230}$Th formed within the waste and then leached out. As seen in Figure 5.1, $N_2 (0, t)$, the concentration of $^{230}$Th at $z = 0$, increases with time.

$^{226}$Ra has been migrating from the waste location just after the beginning of leaching, and its leading edge has arrived at the location $z = v_3 t$.

As shown by the broken lines, the effect of the larger dispersion coefficient ($D = 1 \times 10^3 \text{ m}^2/\text{yr}$) is to smooth the concentration profiles of $^{234}$U and $^{230}$Th, and the gradient discontinuity of $^{230}$Th at $z = v_2 t$ disappears. However, dispersion hardly affects the leading edge and the maximum of the $^{226}$Ra profile, because $^{226}$Ra has a much larger velocity along the migration path, and its decay constant $\lambda_3$ is relatively large.

Figures 5.4 and 5.5 show the concentration profiles of the nuclides after the end of leaching. Before the time of $t=2 \times 10^5$ year, the time of maximum inventory of $^{226}$Ra in Figure 5.1, the total amount of $^{226}$Ra
migrating through the porous medium continues to increase. Since the width of the $^{226}$Ra concentration profile increases with time, and the shape of the profile also changes with time, the highest value of the peak concentration of $^{226}$Ra does not appear at $t=2\times10^5$ yr.

Figure 5.4 shows the concentration profiles at the time $t=5\times10^4$ yr, when the trailing edge of $^{234}$U-band catches up with the first $^{230}$Th from the repository. $^{230}$Th behind the trailing edge of $^{234}$U must then migrate with no further contribution from the decaying $^{234}$U-band, and the amount of $^{230}$Th in this region decreases with time by its own decay. The distribution of $^{226}$Ra is almost completely controlled by $^{226}$Ra which has been produced by the decay of $^{230}$Th leached from the waste.

In Figure 5.5 at $t=1\times10^5$ yr, another peak of $^{230}$Th is growing around the trailing edge of $^{234}$U. The peak of $^{226}$Ra at about 800 m is still increasing compared to that of Figure 5.4. Figure 5.6 shows the concentration profiles of the nuclides at the time $t=2\times10^5$ yr, when the overall total amount of $^{226}$Ra is a maximum. However, the height of the $^{226}$Ra peak is lower than that shown in Figure 5.5. Thus, for the parameters considered here, the global maximum concentration of $^{226}$Ra in the porous medium occurs earlier than the time of maximum inventory of $^{226}$Ra.

The two peaks of $^{230}$Th have almost equal height. The time of $2\times10^5$ yr is around the half-life of $^{234}$U, at which the concentration of $^{234}$U is about half of its initial value.
5.5.2.3 Concentration profiles of nuclides in the case of transient equilibrium of $^{234}\text{U}$ daughters in the waste at $t = 0$.

In the case of initially transient equilibrium of $^{234}\text{U}$ daughters in the waste the relatively large initial amounts of $^{230}\text{Th}$ and $^{226}\text{Ra}$ will have considerable effect on the concentration profiles for migration times less than a few hundred thousand years, i.e., less than a few half lives of $^{230}\text{Th}$.

In the transient equilibrium case, the total amount of nuclides involved in the repository decreases monotonically with time. We would then expect that the maximum concentration of $^{226}\text{Ra}$ along the migration path will occur at an earlier time in comparison with the case of an initially pure $^{234}\text{U}$ source in the repository. This increase of peak $^{226}\text{Ra}$ concentration with distance from the waste, for initial transient equilibrium in the waste, is due entirely to the "reconcentration effect" (B4).

Figure 5.7 shows the concentration profiles of the nuclides at a time before the end of leaching. In the present case of $T = 3 \times 10^4$ yr, the total amount of $^{226}\text{Ra}$ outside the repository increases with time until the end of leaching. As seen in the figure, for the case of small dispersion coefficient, the highest concentration of $^{226}\text{Ra}$ is realized at $z = v_2 t$, i.e., at the location of the first $^{230}\text{Th}$ leached the waste. Because of the large concentration of $^{230}\text{Th}$ between the waste and $z = v_2 t$, and the condition that $v_2 < v_3$, a small peak in the profile of $^{226}\text{Ra}$ occurs. The contribution of $^{230}\text{Th}$ to the concentration
Figure 5.7. Concentration profiles of $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain at $t = 1 \times 10^4$ yr. (Transient equilibrium sources at $t = 0$. $N_i(z,t)$ = atoms/cm$^3$, $N_i^0$ = atoms/cm$^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$.)
profile of $^{226}\text{Ra}$ between the leading edge of $^{234}\text{U}$-band and $z = v_2 t$ is relatively not so large.

The effect of dispersion on the concentration profiles is almost the same as for a pure $^{234}\text{U}$ source, as shown in Figures 5.3 through 5.6. Figures 5.8 through 5.10 show the concentration profiles for times after the end of leaching. The highest peak of $^{226}\text{Ra}$ occurs for the case of small $D$, at the time when the last $^{226}\text{Ra}$ from the waste catches up with the first $^{230}\text{Th}$ nuclide from the waste at $z = v_2 t$. After that, the height of the $^{226}\text{Ra}$ peak decreases with time.

As seen in Figure 5.10, for a transport time of $2 \times 10^5$ yr a second peak of $^{226}\text{Ra}$ concentration is developed at the trailing edge of $^{234}\text{U}$-band. The height of the $^{226}\text{Ra}$ peak at the trailing edge of $^{234}\text{U}$-band decreases more slowly with time than does the peak at $z = v_2 t$, because the half-life of $^{234}\text{U}$ is longer than that of $^{230}\text{Th}$.

5.5.3 Maximum concentration and isopleths of $^{226}\text{Ra}$

Figures 5.11 and 5.12 are the plots of the maximum $^{226}\text{Ra}$ concentration occurring at various locations along the migration path, for the cases of an initially pure $^{234}\text{U}$ source and also for a source with initially transient equilibrium. The two solid curves represent cases of different dispersion coefficients. Each point on the curve means the maximum $^{226}\text{Ra}$ concentration an observer will eventually experience by sitting long enough at a given location. Thus, the time at which the maximum concentration occurs varies with location.
Figure 5.8. Concentration profiles of $^{234}U \rightarrow ^{230}Th \rightarrow ^{226}Ra$ decay chain at $t = 5 \times 10^4$ yr. (Transient equilibrium sources at $t = 0$. $N_i(z,t)$ = atoms/cm$^3$, $N_i^0$ = atoms/cm$^3$ of $^{234}U$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
Figure 5.9. Concentration profiles of $^{234}$U + $^{230}$Th → $^{226}$Ra decay chain at $t = 1 \times 10^5$ yr. (Transient equilibrium source at $t = 0$. $N_i(z,t) = \text{atoms/cm}^3$, $N_i^0 = \text{atoms/cm}^3$ of $^{234}$U at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
Figure 5.10. Concentration profiles of $^{234}\text{U} \rightarrow 230\text{Th} \rightarrow 226\text{Ra}$ decay chain at $t = 2 \times 10^5$ yr. (Transient equilibrium source at $t = 0$. $N_i(z,t) = \text{atoms/cm}^3$, $N_i^0 = \text{atoms/cm}^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$. )
Figure 5.11. Effect of dispersion coefficient on the maximum concentration of $^{226}\text{Ra}$ for the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain. (Pure $^{234}\text{U}$ source at $t = 0$. $N_i(z, t) =$ atoms/cm$^3$, $N_i^0 =$ atoms/cm$^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^6$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
Figure 5.12. Effect of dispersion coefficient on the maximum concentration of $^{226}$Ra for the $^{234}$U $\rightarrow$ $^{230}$Th $\rightarrow$ $^{226}$Ra decay chain. (Transient equilibrium source at $t = 0$.)

$N_i(z,t) = \text{atoms/cm}^3$, $N_1^0 = \text{atoms/cm}^3$ of $^{234}$U at $z = 0$, $t = 0$, $v = 100 \text{ m/yr}$, $T = 3 \times 10^4 \text{ yr}$, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.}
Each point is obtained by choosing a fixed location \( z \) and computing the \(^{226}\text{Ra} \) curve numerically for various times until a maximum concentration is obtained by plotting the concentration versus time for that fixed location. Each of these curves exhibits local maximum \(^{226}\text{Ra} \) concentration. Starting from the waste source at \( z = 0 \), we see an increase in the local maximum \(^{226}\text{Ra} \) concentration with distance away from the waste source until a certain location is reached where the global maximum \(^{226}\text{Ra} \) concentration occurs. The maximum \(^{226}\text{Ra} \) concentration then decreases with further increases in distance. This happens both for the case of initially pure \(^{234}\text{U} \) source and for initially transient equilibrium in the waste. In particular, for the case of initially transient equilibrium in the source, this growth of the peak \(^{226}\text{Ra} \) concentration with distance is called the "reconcentration phenomenon" (B4). This reconcentration phenomenon occurs also for the two-member decay chain \(^{230}\text{Th} \rightarrow ^{226}\text{Ra} \); since for a three-member chain the uranium is providing an additional source by decaying to \(^{230}\text{Th} \), the explanation of this effect should be similar for both the two-member and three-member chains.

Consider radionuclide propagation for the simple two-member model. When the band of \(^{230}\text{Th} \) reaches an observer near the waste source the observer will detect \(^{226}\text{Ra} \) from the immediate decay of \(^{230}\text{Th} \) at that position. At the same time \(^{226}\text{Ra} \) from the earlier decay of \(^{230}\text{Th} \) nearer the waste source can also reach the observer at that instance. If the observer is far enough from the waste source so that \(^{226}\text{Ra} \) directly from the source decays before reaching the observer, it is the band of \(^{230}\text{Th} \) between the waste and the observer that contributes to the concentration of \(^{226}\text{Ra} \) at the observer's position. When still farther
away from the source, but still not far enough for significant decay of the longer-lived $^{230}\text{Th}$, a longer band of $^{230}\text{Th}$ contributes to the simultaneous arrival of more $^{226}\text{Ra}$ at the observer location; thereby resulting in the increase in the local maximum $^{226}\text{Ra}$ concentration shown in Figures 5-11 and 5-12. When the distance of the observer is sufficiently increased, a longer time is required for $^{230}\text{Th}$ to reach the observer, the $^{230}\text{Th}$ itself will have appreciably decayed, and the local maximum $^{226}\text{Ra}$ concentration begins to decrease with increasing distance from the source. Thus one of the main reasons for this reconcentration phenomenon to occur is the greater migration speed and shorter half life of the daughter nuclide.

In both Figures 5-11 and 5-12 the local maximum $^{226}\text{Ra}$ concentrations are of greater magnitude for the smaller dispersion coefficients. In both Figures 5-11 and 5.12 the locations of the global maximum $^{226}\text{Ra}$ concentration for the initially pure $^{234}\text{U}$ waste are 840 m for $D = 1 \times 10^{-1} \text{m}^2/\text{yr}$ and 800 m for $D = 1 \times 10^{-3} \text{m}^2/\text{yr}$. For initially transient equilibrium the global maxima are at 70 m for $D = 1 \times 10^{-1} \text{m}^2/\text{yr}$ and 86 m for $D = 1 \times 10^{-3} \text{m}^2/\text{yr}$. The magnitude of the global maximum $^{226}\text{Ra}$ concentration is about five-fold greater for initial transient equilibrium in the waste.

The effect of the larger dispersion on the local maximum $^{226}\text{Ra}$ concentration is greater near the waste, which is a consequence of dispersion of $^{230}\text{Th}$ upstream of the waste. This effect is more clearly shown in the isopleths of $^{226}\text{Ra}$ concentration given in Figure 5.13 and 5.14 for the initially pure $^{234}\text{U}$, and in Figure 5.15 and 5.16 for initially transient equilibrium. The abscissa shows distance (km) and
the ordinate shows time (yr). The isopleths are shown for values of \( N_3(z,t)/N_1^0 \) from \( 5 \times 10^{-3} \) to \( 2 \times 10^{-2} \) for each isopleth for the initially pure \( ^{234}\text{U} \) waste, and from \( 2 \times 10^{-2} \) to \( 8 \times 10^{-2} \) for initially transient equilibrium waste.

Let us consider Figures 5.13 and 5.14, which are for \( D = 1 \times 10^{-1} \) and \( D = 1 \times 10^{-3} \text{ m}^2/\text{yr} \). As shown in these figures the effect of dispersion on the slope of the isopleth is less important except near the repository. The broken lines in these figures indicate the time when the concentration of \( ^{226}\text{Ra} \) at any given location reaches its maximum value, thereby defining the ridge line of the isopleths. Each ridge line lies on the line of \( T = 3 \times 10^4 \text{ yr} \) (leach time) near the waste source and up to a distance of about 0.1 km. Finally at large distances the ridge line lies asymptotically along the line \( t = z/v_1 \), which is the locus of the leading edge of \( ^{234}\text{U} \) band.

The location of the ridge line is not much affected by changes in the dispersion coefficient for the case of pure \( ^{234}\text{U} \) source. The effect of dispersion is much more important in the case of initial transient equilibrium, as shown in Figures 5.15 and 5.16. For the small-dispersion case of Figure 5.15, the locus of maximum concentration almost agrees with the line \( t = z/v_2 \), which is the locus of the leading edge of \( ^{230}\text{Th} \), from the waste source up to 60 m distance. Then it lies on the line \( t = 3 \times 10^4 \text{ yr} \) (equal to the leach time) up to \( z = 200 \text{ m} \), and finally it approaches asymptotically the line \( t = z/v_1 \). However, for the large-dispersion case of Figure 5.16, the locus of the maximum concentration does not lie on the line \( t = z/v_2 \) but instead lies on
Figure 5.13. Isopleths of $^{226}\text{Ra}$ for the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain. (Pure $^{234}\text{U}$ source at $t = 0$. $N_i(z, t) =$ atoms/cm$^3$, $N_0 = $ atoms/cm$^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $D = 1 \times 10^{-1}$ m$^2$/yr, $T = 3 \times 10^4$ yr, $K_u = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$. )
Figure 5.14. Isopleths of $^{226}\text{Ra}$ for the $^{234}\text{U} \rightarrow ^{230}\text{Th} + ^{226}\text{Ra}$ decay chain. (Pure $^{234}\text{U}$ source at $t = 0$. $N_i(z,t)$ = atoms/cm$^3$, $N_i^0$ = atoms/cm$^3$ of $^{234}\text{U}$ at $z = 0$, $v = 100$ m/yr, $D = 1 \times 10^3$ m$^2$/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
Figure 5.15. Isopleths of $^{226}\text{Ra}$ for the $^{234}\text{U} \to ^{230}\text{Th} \to ^{226}\text{Ra}$ decay chain. (Transient equilibrium sources at $t = 0$.

\[ N_i(z,t) = \text{atoms/cm}^3, \quad N_1^0 = \text{atoms/cm}^3 \text{ of } ^{234}\text{U} \text{ at } z = 0, \]

\[ t = 0, \quad v = 100 \text{ m/yr}, \quad D = 1 \times 10^{-1} \text{ m}^2/\text{yr}, \quad T = 3 \times 10^4 \text{ yr}, \]

\[ K_U = 1 \times 10^4, \quad K_{\text{Th}} = 5 \times 10^4, \quad K_{\text{Ra}} = 5 \times 10^{-2}. \]
Figure 5.16. Isopleths of $^{226}_{\text{Ra}}$ for the $^{234}_{\text{U}} \rightarrow ^{230}_{\text{Th}} \rightarrow ^{226}_{\text{Ra}}$ decay chain. (Transient equilibrium sources at $t = 0$. $N_1(z,t) = \text{atoms/cm}^3$, $N_1^0 = \text{atoms/cm}^3$ of $^{234}_{\text{U}}$ at $z = 0$, $t = 0$, $v = 100 \text{ m/yr}$, $D = 1 \times 10^3 \text{ m}^2/\text{yr}$, $T = 3 \times 10^4 \text{ yr}$, \( K_U = 1 \times 10^4 \), \( K_{\text{Th}} = 5 \times 10^4 \), \( K_{\text{Ra}} = 5 \times 10^2 \).)
the leach-time line of $t = 3 \times 10^4$ yr from the waste source up to a distance of $z = 240$ m. This behavior may come from the shape of the edge of the $^{230}$Th band at $t = z/v_2$, as shown in Figures 5.7-5.10. For the small-D case, the $^{230}$Th concentration profile is very steep at this point, resulting in a maximum $^{226}$Ra concentration at that same position. For the large-D case, the $^{226}$Ra peak is shifted to $t > z/v_2$ because the sharp edge of $^{230}$Th band no longer exists.

5.5.4 Parametric study

5.5.4.1 The effect of the sorption retardation coefficient on the concentration profiles of $^{226}$Ra

Figure 5.17 shows the concentration profiles of $^{226}$Ra at various locations for different sorption retardation coefficients for $^{226}$Ra ($K_3/10, K_3, 10K_3$), for an initially pure $^{234}$U source, at $t = 5 \times 10^4$ yr and with $D = 1 \times 10^{-1}$ m$^2$/yr. In the figure, the abscissa is a linear scale in units of $zK_3/v$ years, and the ordinate is the relative concentration of $^{226}$Ra. The assumed value of $K_3$ is 400, that was used in the calculations previously discussed. The $K_3$ and $K_3/10$ curves decrease with $zK_3/v$ linearly, and their slopes are $-4.69 \times 10^{-4}$ yr$^{-1}$ for the $K_3$ curve, and $-4.33 \times 10^{-4}$ yr$^{-1}$ for the $K_3/10$ curve. Since the decay constant of $^{226}$Ra has the value of $4.332 \times 10^{-4}$ yr$^{-1}$, the same as the value of the slope for the $K_3/10$ curve, we find that at a distance from the repository requiring about two or more years for $^{226}$Ra transport time, the concentration of $^{226}$Ra decreases exponentially according to $e^{-\lambda_3 zK_3/v}$, provided the migration speed of $^{226}$Ra is much greater ($K_2/K_3 \sim 100$) than that of its parent nuclide. This characteristic can be explained by the explicit solution for the dispersion-free
Figure 5.17. Effect of sorption retardation constant on concentration profiles of $^{226}$Ra at $t = 5 \times 10^4$ yr for the $^{234}$U + $^{230}$Th + $^{226}$Ra decay chain. (Pure $^{234}$U source at $t = 0$. $N_i(z,t) = \text{atoms/cm}^3$, $N_i^0 = \text{atoms/cm}^3$ of $^{234}$U at $z = 0$, $t = 0$, $v = 100$ m/yr, $D = 1 \times 10^{-1}$ m$^2$/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 4 \times 10^2$.)
case and a step release, Eq. (5.16). For \( v_1 t < z < v_3 t \), the solution is as follows:

\[
N_3^S(z, t) = e^{-\frac{\lambda_3 z}{v_3}} \left[ b_{31} e^{-\lambda_1 (t-z/v_3)} + b_{32} e^{-\lambda_2 (t-z/v_3)} + b_{33} e^{-\lambda_3 (t-z/v_3)} \right] \\
+ \frac{\lambda_1 \lambda_2}{v_1 v_2} \frac{\lambda_3}{r_{12}} \left[ d_{13} b_{11} \left\{ e^{-\Delta_{13} (t-z/v_3)} - e^{-\lambda_1 (t-z/v_3)} \right\} \\
+ d_{23} b_{21} \left\{ e^{-\Delta_{23} (t-z/v_3)} - e^{-\lambda_1 (t-z/v_3)} \right\} \\
+ d_{33} b_{31} \left\{ e^{-\Delta_{33} (t-z/v_3)} - e^{-\lambda_1 (t-z/v_3)} \right\} \right] \\
+ \frac{\lambda_2}{v_2} \frac{\lambda_3}{r_{23}} \left[ d_{23} b_{21} \left\{ e^{-\Delta_{23} (t-z/v_3)} - e^{-\lambda_2 (t-z/v_3)} \right\} \\
+ d_{33} b_{31} \left\{ e^{-\Delta_{33} (t-z/v_3)} - e^{-\lambda_2 (t-z/v_3)} \right\} \right] \\
(5.44)
\]

Let's consider the case that \( \lambda_3 \gg \lambda_1, \lambda_2; v_3 \gg v_1, v_2 \) and \( t \gg 1/\lambda_3 \). These conditions yield that \( \lambda_3 - \lambda_i \approx \lambda_3, v_3 - v_1 \approx v_3 \) and \( e^{-\lambda_3 t} \approx 0 \) where \( i = 1 \) or \( 2 \). With this approximation, one can rewrite (5.44) as

\[
N_3^S(z, t) \approx G(t) e^{-\frac{\lambda_3 z}{v_3}} \]

(5.45)

where
\[ G(t) = B_{31} e^{-\lambda_1 t} + B_{32} e^{-\lambda_2 t} \]
\[ + \frac{\lambda_1 \lambda_2}{\nu_1 \nu_2} \frac{1}{\Gamma_{13} \Gamma_{23}} \left[ \frac{D_{31} B_{11}}{\lambda_1} \left( e^{-\Delta_{13} t} - e^{-\lambda_1 t} \right) + \frac{D_{31} B_{12}}{\lambda_1 \Delta_{23}} \left( e^{-\lambda_2 t} - e^{-\lambda_1 t} \right) \right] \]
\[ + \frac{\lambda_2}{\nu_2} \frac{1}{\Gamma_{23}} \left[ \frac{D_{32} B_{21}}{\lambda_1 \Delta_{23}} \left( e^{-\Delta_{23} t} - e^{-\lambda_1 t} \right) + \frac{D_{32} B_{22}}{\lambda_2 \Delta_{23}} \left( e^{-\lambda_2 t} - e^{-\lambda_2 t} \right) \right] \]

(5.46)

Using the superposition relation (3.60), one can obtain the solution of \(^{226}\text{Ra}\) concentration, \(N^b_3(z,t)\), for the band release as

\[ N^b_3(z,t) = N^g_3(z,t; B_{ij}) - N^g_3(z,t-T; B_{ij} e^{-\lambda_j T}) \]
\[ = \left[ G(t; B_{ij}) - G(t-T; B_{ij} e^{-\lambda_j T}) \right] e^{-\lambda_3 z/\nu_3} \]

(5.47)

Thus, the solution for the band release shows that the concentration of \(^{226}\text{Ra}\) decreases with distance exponentially according to \(e^{-\lambda_3 z/\nu_3}\), where \(\nu_3 = \nu/K_3\).

5.5.4.2 The effect of sorption retardation coefficient on the maximum discharge rate of \(^{226}\text{Ra}\)

Figures 5.18 and 5.19 show the effect of the local maximum discharge rate of \(^{226}\text{Ra}\), for \(D = 1 \times 10^{-1} \text{ m}^2/\text{yr}\) and \(1 \times 10^3 \text{ m}^2/\text{yr}\), for an initially pure \(^{234}\text{U}\) source, and for a leach time of \(3 \times 10^4 \text{ yr}\). The maximum discharge rate at a given location is the product of the maximum concentration at that location and the groundwater flow rate. Here the amount of waste material is that in the high-level reprocessing wastes formed.
Figure 5.18. Effect of sorption retardation constant on maximum discharge rate of $^{226}\text{Ra}$ for the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain, with $D = 1 \times 10^{-1}$ m$^2$/yr. (Pure $^{234}\text{U}$ source at $t = 0$.

$N^1(t, x) = \text{atoms/cm}^3$, $N^0 = \text{atoms/cm}^3$ of $^{234}\text{U}$ at $z = 0$,

t = 0, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
Figure 5.19. The effect of sorption retardation constant on maximum discharge rate of $^{226}$Ra for the $^{234}$U $\rightarrow$ $^{230}$Th $\rightarrow$ $^{226}$Ra decay chain with $D = 1 \times 10^3$ m$^2$/yr. (Pure $^{234}$U source at $t = 0$. $N_i(z,t)$ = atoms/cm$^3$, $N_i^0$ = atoms/cm$^3$ of $^{234}$U at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$/yr, $K_U = 1 \times 10^4$, $K_{TH} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.)
by operating a light-water power reactor for 1 GWe-yr. \textsuperscript{238}Pu, \textsuperscript{242m}Am, and \textsuperscript{242}Cm in the waste is assumed to have decayed to \textsuperscript{234}U before a waste dissolution begins. Since a groundwater flow rate associated with this amount of waste must be assumed to calculate an actual concentration of dissolved radionuclides, the assumed water flow rate cancels when calculating the discharge rate. The resulting initial activity of \textsuperscript{234}U is estimated to be 7.67 Ci/GWe-yr. The values of K\textsubscript{3} assumed (5, 50, 500, 5000) are all smaller than that of K\textsubscript{2} (50,000), so the local maximum discharge rate of \textsuperscript{226}Ra reaches a global maximum at a location away from the repository. The magnitude of the global maximum discharge rate and the distance over which appreciable discharge occurs both increase as K\textsubscript{3} decreases. However, reducing the radium sorption-retardation coefficient below the presently assumed value of 500 results in only a relatively small increase in the maximum discharge rate.

Comparison of Figures 5.18 and 5.19 with regard to different dispersion coefficient shows that the effect of dispersion on the curves of \textsuperscript{226}Ra discharge, for K\textsubscript{3}, K\textsubscript{3}/10 and K\textsubscript{3}/100, is slight except near the waste source. This is explained by the fact that for small K\textsubscript{3}, i.e., for large radium migration speed, the transport of \textsuperscript{226}Ra is controlled more by convection than by dispersion.
5.5.4.3 Effect of leach time on the maximum discharge rate of $^{226}\text{Ra}$

Figures 5.20 and 5.21 show the effect of varying leach time T on the maximum discharge rate of $^{226}\text{Ra}$ at various locations for an initially pure $^{234}\text{U}$ source and for dispersion coefficients of $1 \times 10^{-1}$ and $1 \times 10^{3}$ m$^2$/yr. These figures show that the global maximum discharge rates, and the locations of the global maximum, are not significantly affected by a ten-fold increase in leach time from $3 \times 10^3$ yr to $3 \times 10^4$ yr. Even a hundred-fold increase in leach time $3 \times 10^3$ yr to $3 \times 10^5$ yr causes only a two-fold reduction in the global maximum rate of radium discharges. The leach-time effects are similarly weak at distances greater than that at which the global maximum occurs.

Nearer the repository increasing the leach time from $3 \times 10^3$ yr to $3 \times 10^4$ yr increases the local maximum discharge rate of $^{226}\text{Ra}$. The longer leach time allows more $^{230}\text{Th}$ and $^{226}\text{Ra}$ to form in the waste prior to complete dissolution, thereby increasing the amount of those nuclides in the near field of the waste source as leaching continues. For the even higher leach time of $3 \times 10^5$ yr there is little further ingrowth of $^{230}\text{Th}$ in the waste, so the net effect of the lower dissolution rate is to reduce the near-field concentrations of $^{230}\text{Th}$ and $^{226}\text{Ra}$.

Figure 5.22 shows the dependence of the global maximum discharge rate of $^{226}\text{Ra}$ on leach time, for an initially pure $^{234}\text{U}$ source and for initially a $^{234}\text{U}$ source with $^{230}\text{Th}$ and $^{226}\text{Ra}$ in transient equilibrium. For the initially pure $^{234}\text{U}$ source there is little effect of leach time, for leach times below $10^5$ yr, as explained above. For a waste source in initial transient equilibrium, the global maximum discharge rate
Figure 5.20. Effect of leach time of the maximum discharge rate of $^{226}\text{Ra}$ for the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain, with

$D = 1 \times 10^{-1}$ m$^2$/yr. (Pure $^{234}\text{U}$ source at $t = 0$. $N_i(z,t)$ =

atoms/cm$^3$, $N_i^0$ = atoms/cm$^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$. )
Figure 5.21. Effect of leach time on the maximum discharge rate of $^{226}\text{Ra}$ for the $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ decay chain, with $D = 1 \times 10^3 \text{ m}^2/\text{yr}$. (Pure $^{234}\text{U}$ source at $t = 0$. $N_1(z,t) = \text{atoms/cm}^3$, $N_1^0 = \text{atoms/cm}^3$ of $^{234}\text{U}$ at $z = 0$, $t = 0$, $v = 100 \text{ m/yr}$, $T = 3 \times 10^4 \text{ yr}$, $K_U = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$.)
Figure 5.22. Effect of leach time on the highest concentration of $^{226}$Ra for the $^{234}$U $\rightarrow$ $^{230}$Th $\rightarrow$ $^{226}$Ra decay chain. $N_i(z,t)$ = atoms/cm$^3$. $N_i^0$ atoms/cm$^3$ of $^{234}$U at $z = 0$, $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_U = 1 \times 10^4$, $K_{Th} = 5 \times 10^4$, $K_{Ra} = 5 \times 10^2$.
of $^{226}\text{Ra}$ varies inversely with leach time except in the region of a leach time of $10^4$ yr.

If water intrusion and waste corrosion occur within a few tens of thousands of years after emplacement, the waste will contain little of the $^{234}\text{U}$ daughters at leach resistance the beginning of dissolution. Increasing the corrosion resistance of the waste does not reduce the local maximum discharge rate of $^{226}\text{Ra}$, unless extremely low corrosion rates, corresponding to leach times of millions of years, are achieved and demonstrated. This conclusion may be important to these analyses of the long-term environmental effects of waste dissolution and radio-nuclide transport wherein dose rates from $^{226}\text{Ra}$ reaching the biosphere have been found to be important and, in some cases, controlling.

5.6 Migration Behavior of the 4n+1 Decay Chain

The members of the 4n+1 ($n = 1, 2, 3, \ldots$) decay chain which may occur in high-level radioactive wastes are:

\[
^{241}\text{Pu} \rightarrow ^{241}\text{Am} \rightarrow ^{237}\text{Np} \rightarrow ^{233}\text{Pa} \rightarrow ^{233}\text{U} \rightarrow ^{229}\text{Th} \rightarrow ^{225}\text{Ra} \rightarrow
\]

The first three members are present in unprocessed spent fuel and in high-level reprocessing waste.

5.6.1 Reduction to a three-member decay chain

We will assume that relatively little corrosion of the radioactive waste occurs for several hundred years after emplacement in a repository. Therefore, we may assume that all $^{241}\text{Pu}$ and $^{241}\text{Am}$ have decayed to $^{237}\text{Np}$ before dissolution begins. The half-lives, assumed sorption retardation constants (B3), elemental migration speed, and nuclide migration distance
for the time of a half-life for $^{237}$Np and its decay daughters are shown in Table 5.3.

As seen in the above table, $^{233}$Pa and $^{225}$Ra have very short half-lives, and the distances of migration in their half-lives, are very short in comparison with those for the other three nuclides. Therefore, as discussed in Section 4.5, $^{233}$Pa and $^{225}$Ra are expected to be in secular equilibrium with each of their parents, $^{237}$Np and $^{229}$Th, at every location and at any time. Therefore, we can reduce this five-member decay chain to the three-member decay chain: $^{237}$Np $\rightarrow$ $^{233}$U $\rightarrow$ $^{229}$Th.

At any location within the sorbing medium the radioactivity level of $^{225}$Ra in groundwater will be related to the radioactivity level of $^{229}$Th by Eq. (4.134).

5.6.2 Discharge-rate profiles of the $^{237}$Np $\rightarrow$ $^{233}$U $\rightarrow$ $^{229}$Th ($\rightarrow$ $^{225}$Ra) decay chain

Figure 5.23 shows the space dependence of the discharge rate of each nuclide at $t = 5 \times 10^4$ yr, for a leach time of $3 \times 10^4$ yr, and for an initially pure $^{237}$Np source. The basis for calculating these discharge rates is the same as explained in Section 5.5.4.2. An estimation of the initial activity of $^{237}$Np in high-level reprocessing wastes from a light-water power reactor is 14.4 Ci/GWe-yr (B2). $^{233}$Pa and $^{225}$Ra are in secular equilibrium with $^{237}$Np and $^{229}$Th, respectively. The curves for $^{229}$Th are identical with those for $^{225}$Ra. This figure shows also the effect of varying the dispersion coefficient.

$^{237}$Np migrates relatively fast in this three member chain. The migration speeds are in the order of $v_1 > v_2 > v_3$. Thus, the first two nuclides have the tendency to leave their daughters behind. The
<table>
<thead>
<tr>
<th></th>
<th>$^{237}$Np</th>
<th>$^{233}$Pa</th>
<th>$^{233}$U</th>
<th>$^{229}$Th</th>
<th>$^{225}$Ra</th>
</tr>
</thead>
<tbody>
<tr>
<td>Half-life, $T_{1/2}$ (yr)</td>
<td>$2.14 \times 10^6$</td>
<td>$7.4 \times 10^{-2}$</td>
<td>$1.62 \times 10^5$</td>
<td>$7.34 \times 10^3$</td>
<td>$4.1 \times 10^{-2}$</td>
</tr>
<tr>
<td>Retardation coefficient, $K_i^a$</td>
<td>$1 \times 10^2$</td>
<td>$1.7 \times 10^4$</td>
<td>$1 \times 10^4$</td>
<td>$5 \times 10^4$</td>
<td>$5 \times 10^2$</td>
</tr>
<tr>
<td>Elemental migration speed, $v_i$ (m/yr)$^b$</td>
<td>$1 \times 10^0$</td>
<td>$6 \times 10^{-3}$</td>
<td>$1 \times 10^{-2}$</td>
<td>$2 \times 10^{-3}$</td>
<td>$2 \times 10^{-1}$</td>
</tr>
<tr>
<td>$v_i \times T_{1/2}$ (m)</td>
<td>$2.14 \times 10^6$</td>
<td>$4.44 \times 10^{-4}$</td>
<td>$1.62 \times 10^3$</td>
<td>$1.47 \times 10^1$</td>
<td>$8.2 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

$^a$ Assumed values, from (B3)

$^b$ Assuming $v = 100$ m/yr
Figure 5.23. Concentration profiles of $^{225}\text{Ra}$ of $^{237}\text{Np} + ^{233}\text{Pa} + ^{233}\text{U} + ^{229}\text{Th} + ^{225}\text{Ra}$ decay chain at $t = 5 \times 10^4$ yr. (Pure $^{237}\text{Np}$ source at $t = 0$. $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_{\text{Np}} = 10^2$, $K_{\text{Pa}} = 1 \times 10^4$, $K_{\text{U}} = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$.)
dip of $^{225}$Ra between $v_3 t$ and $v_2 (t - T)$ in the curve for $D = 1 \times 10^{-1} m^2/yr$ is due to the competition between the increase with time in the concentration of $^{229}$Th in the undissolved waste and the decay of $^{229}$Th lagging behind its parent nuclide $^{233}$U. The effect of the larger dispersion is to smooth and spread out the profiles of each nuclide.

Figure 5.24 is the plot of the local maximum discharge rate of $^{225}$Ra versus location, under the same conditions as those of Figure 5.23. The maximum discharge rate decreases monotonously with distance from the waste source. The low-dispersion curve can be discussed in three separate regions along the migration path. In the first region, $z < \sim 0.2 \text{ km}$, the maximum discharge rate at any given location happens to occur when the last $^{229}$Th produced inside the waste passes that location. In the second region, $0.2 \text{ km} < z < 9 \text{ km}$, the maximum discharge rate of $^{255}$Ra occurs at the same instant that the peak of $^{233}$U occurs. In the last region, $z < 9 \text{ km}$, the maximum $^{225}$Ra discharge rate appears at $z/v_1 + 1.7 \times 10^5 \text{ yr}$. This constant local-maximum discharge rate extends far from the waste source because the first nuclide, $^{237}$Np, has a relatively large migration speed and a long half life. Reconcentration (see Sect. 5.5.3) of $^{225}$Ra does not occur at any location because the assumed sorption retardation constant of $^{229}$Th is larger, and its migration speed smaller, than that of its parent nuclides.
Figure 5.24. Effect of dispersion coefficient on maximum discharge rate of $^{225}\text{Ra}$ for the $^{237}\text{Np} \rightarrow ^{233}\text{Pu} \rightarrow ^{229}\text{Th} \rightarrow ^{225}\text{Ra}$ decay chain. (Pure $^{237}\text{Np}$ source at $t = 0$, $v = 100$ m/yr, $T = 3 \times 10^4$ yr, $K_{\text{Np}} = 10^2$, $K_{\text{Pa}} = 1 \times 10^4$, $K_{\text{U}} = 1 \times 10^4$, $K_{\text{Th}} = 5 \times 10^4$, $K_{\text{Ra}} = 5 \times 10^2$.)
6. NUCLIDE MIGRATION IN A MULTI-LAYERED GEOLOGIC MEDIUM

6.1 Introduction

This chapter deals with the effect of a multi-layer system on nuclide migration. It seems likely that the nuclides will encounter geologic media of different properties before reaching the biosphere. These media may be characterized by different values of:

\[ v = \text{water velocity} \]
\[ K_i = \text{the sorption retardation coefficient} \]
\[ S_f = \text{the cross sectional area of the water phase} \]
\[ \varepsilon = \text{the porosity} \]
\[ D_i = \text{the dispersion coefficient, etc.} \]

Each layer is supposed to have constant properties of \( K_i, S_f, \varepsilon, \) and \( D_i \), and for this one-dimensional analysis the water velocity \( v \) is assumed to be constant within a layer. The transport equation discussed in the preceding chapters is valid in each layer. The boundary condition at the interface between two layers is given by the continuity of nuclide flux and nuclide concentration. For the case of no axial dispersion, a recursive solution can be easily found by the use of the general solution for a single-medium system (see Chapter 4). Some applications will be given there for a two-medium system:

a) A general formula, for an arbitrary release mode at the repository, and an i-member chain,
b) A general formula for a step release and an i-member chain,
c) A step-release formula for the first, second and third members
of a radioactive decay chain.

The solution for a band release can be found by the superposition principle,
which has been derived in Chapter 3.

For the case with axial dispersion, a recursive formula is given
for a two-medium system and a generalized release mode at the repository.

6.2 Transport Equation and Conditions at the Layer Boundary

In this chapter we consider the case of a multilayer system. Each
layer is characterized by the superscript, \( \ell \), at the left, e.g.,

\[ \ell N_i(z,t) = \text{concentration of nuclide } i, \text{ at } z \text{ and } t \text{ in the } \ell^{th} \text{ layer}, \]

\[ \ell D_i = \text{axial dispersion coefficient of nuclide } i, \text{ in the } \ell^{th} \text{ layer}, \]

\[ \ell v = \text{water velocity in the } \ell^{th} \text{ layer}, \text{ and so on.} \]

We denote the position of the interface between the \( \ell^{th} \) and the \((\ell + 1)^{th}\)
layers by \( \ell z \).

\[ \begin{align*}
  &1_z &2_z &3_z &4th \text{ layer} \\
  &1st \text{ layer} &2nd \text{ layer} &3rd \text{ layer} &
\end{align*} \]

Figure 6.1. Multi-layer Pathway.
We assume the following parameters to be constant in each layer:

\[ \ell_{D_i}, \ell_{K_i}, \ell_{v_i}, \ell_v, \ell_c, \ell_{S_f}, \ell_{S_s} \]

Under these assumptions the transport equation, as discussed in Chapter 2 is still valid in each layer. The transport equation in the \( \ell \)th layer can be written as:

\[ \ell_{L_i}^{\ell} N_i(z,t) = \ell_{K_i-1} \ell_{i-1} \ell_{N_i-1} \quad ; \quad \ell-1 \leq z \leq \ell z \quad (6.1) \]

where the operator \( \ell_{L_i}^{\ell} \) is defined by

\[ \ell_{L_i}^{\ell} \equiv -D_i \frac{\partial^2}{\partial z^2} + v \frac{\partial}{\partial z} + K_i \frac{\partial}{\partial t} + \frac{\partial}{\partial t} \quad (6.2) \]

At the interface \( \ell z \) we must have the continuity of the nuclide flux, i.e., the rate at which nuclide \( i \) leaves the layer \( \ell \) equals the rate at which it enters the layer \( \ell+1 \):

\[ \ell_{S_f} \left[ -\ell_{D_i} \frac{\partial N_i}{\partial z} + \ell_v \ell_{N_i} \right]_{z=\ell z} = \ell+1_{S_f} \left[ -\ell+1_{D_i} \frac{\partial^{\ell+1} N_i}{\partial z} + \ell+1_v \ell^{\ell+1} N_i \right]_{z=\ell z} \quad (6.3) \]

The term \( -D_i \frac{\partial N_i}{\partial z} \) represents the transport by diffusion and the term \( -v \ell_{N_i} \) by convection. By introducing

\[ \ell_{d_i} = \frac{\ell_{D_i}}{\ell_v} \quad (6.4) \]
and by the use of the conservation of mass of water,

\[ \lambda S_f \lambda \nabla \rho = \lambda + 1 \lambda S_f \lambda + 1 \lambda \nabla \rho \]  \hspace{1cm} (6.5)

we can rewrite (6.3) as

\[ \lambda I \left[ - \lambda d_i \frac{\partial N_i}{\partial z} + \lambda N_i \right] = - \lambda + 1 \lambda d_i \frac{\partial^{\lambda + 1} N_i}{\partial z} + \lambda + 1 \lambda N_i \hspace{1cm} \text{at } z = \ell \]  \hspace{1cm} (6.6)

where

\[ \lambda I = \frac{\lambda + 1 \lambda I}{\lambda \rho} \]  \hspace{1cm} (6.7)

In most cases, the water density can be considered a constant, therefore the term \( \lambda I \) can be set equal to unity. Since transport by diffusion is independent of transport by convection, (6.6) can be split into two equations, and we obtain (with \( \lambda I = 1 \)):

\[ \lambda N_i = \lambda + 1 N_i \hspace{1cm} \text{at } z = \ell \]  \hspace{1cm} (6.8)

and

\[ \lambda d_i \frac{\partial N_i}{\partial z} = \lambda + 1 \lambda d_i \frac{\partial^{\lambda + 1} N_i}{\partial z} \hspace{1cm} \text{at } z = \ell \]  \hspace{1cm} (6.9)

6.3 Analytical Solutions for the Non-Dispersion Case

In the absence of dispersion, the transport equation becomes:

\[ \lambda \frac{\partial N_i}{\partial z} + K_i \frac{\partial N_i}{\partial t} + K_i \lambda_i N_i = K_{i-1} \lambda_i \lambda N_i - 1 \hspace{1cm} \lambda - 1 \leq z \leq \ell \]  \hspace{1cm} (6.10)
with

I.C. \( \frac{\partial}{\partial z} N_i(z,0) = 0 \); all l's \hspace{1cm} (6.11) 

B.C. \( N_i(0,t) = \phi_i(t) \); at the waste source \hspace{1cm} (6.12) 

\[ \frac{\partial}{\partial z} N_i(\ell_{-1}z, t) = N_i(\ell_{-1}z, t) \] \hspace{1cm} (6.13) 

We define for each interval \( \ell \) the new variable:

\[ \ell_{\sim} z \equiv z - \ell_{-1} z \] \hspace{1cm} (6.14)

In terms of this variable, we have now the problem:

Solve

\[ \frac{\partial}{\partial z} N_i(\ell_{\sim}z, t) + \frac{\partial}{\partial t} N_i(\ell_{\sim}z, t) + \frac{\partial}{\partial \lambda_{i-1}} N_i(\ell_{\sim}z, t) = \lambda_{i-1} N_i(\ell_{\sim}z, t) \] \hspace{1cm} (6.15)

with

I.C. \( \frac{\partial}{\partial z} N_i(\ell_{\sim}z,0) = 0 \); all l's \hspace{1cm} (6.16) 

B.C. \( N_i(0,t) = \phi_i(t) \); all l's \hspace{1cm} (6.17) 

where \( \phi_i(t) \) is given by (6.12) and

\[ \phi_i(t) = \frac{\partial}{\partial t} \frac{\partial}{\partial \lambda_{i-1}} N_i(\ell_{-1}z, t) ; \ell \geq 2 \] \hspace{1cm} (6.18) 

and

\[ \phi_i(t) = 0 ; \ t < 0, \ \text{all l's} \]
If (6.15), (6.16) and (6.17) are compared with the problem posed in Chapter 4.42 we see that we have here the same problem. We can therefore use the general solution (4.127) shown in 4.42 for each layer. For clarity, denote the solution by $\mathcal{N}_i(z, t; \lambda \phi_i(t))$.

$$\mathcal{N}_i(z, t; \lambda \phi_i(t)) \equiv e^{-\left(\lambda \beta_{v_i}\right)z} \phi_i(t-z/\beta_{v_i})$$

$$+ \sum_{j=1}^{i-1} \sum_{m=j}^{1} \sum_{r,m} \frac{1}{\beta_{v_i} \beta_{v_i}^{(1)}} e^{-\left(\lambda \beta_{v_i}\right)z} \sum_{r,m} \beta_{v_i}^{(1)} \beta_{v_i}^{(2)}(z, t) \phi_j(t)$$

(6.19)

where $g \otimes \phi$ represents the convolution product and $\lambda \phi_i(t)$ represents the dependence on $\lambda \phi_i(t), \ldots \lambda \phi_i(t)$. The other symbols are the ones used in Chapter 4. The superscript, $\lambda$, means that we must take $\lambda \nu_i = \nu_i / \nu_i$ whenever the values $\nu_i$ appears. The solution for each layer is then:

$$\mathcal{N}_i(z, t) = \mathcal{N}_i(z, \lambda \phi_i(t)); 0 \leq z \leq \lambda^{-1}$$

(6.20)

the $\lambda \phi_i$'s being given by (6.18).

Finally, in terms of $z$ we have

$$\mathcal{N}_i(z, t) = \mathcal{N}_i(z^{-1}, \lambda \phi_i(t)); \lambda^{-1} \leq z \leq \lambda$$

(6.21)

where $\lambda \phi_i(t)$ is given by the nuclide release mode at the repository and

$$\lambda \phi_i(t) \equiv \frac{\lambda \rho}{\lambda^{-1} \rho} \mathcal{N}_i(\lambda^{-1} z, t) \quad \text{for } \lambda \geq 2$$

---

\footnote{The parameter $\mathcal{N}_i^0$, which appeared explicitly in the previous chapters has now been assimilated with the symbol $\lambda \phi_i(t)$.}
Equation (6.21) is a "doubly recursive" solution, i.e., it is recursive both in space and in the nuclides. One must calculate first $^1N_j(z,t)$ for $j = 1,2,3,...i$, using equation (6.21), then calculate $^2N_j(z,t)$ for $j = 1,2,...i$, using the previous results and (6.21), and so on up to $^iN_i(z,t)$.

6.4 Application of the Recursive Solution to a Two-Medium System

6.4.1 General release mode at the repository: $^1\phi_i(t)$

We have:

$$^1N_i(z,t) = ^1\mathcal{N}_i(z,t; ^1\phi_i(t))$$

(6.22)

and (6.21) yields:

$$^2N_j(z,t) = e^{-\lambda_i(z-\frac{1}{2})} \frac{r_0}{l_\rho} \left\{ e^{-\lambda_j(z-\frac{1}{2})^2} e^{\frac{1}{2}(z-\frac{1}{2})^2} \left[ b_{1,0} \right] + \sum_{j=1}^{i-1} A_{i,j} \sum_{m=j}^{i-1} b_{m,j-m} e^{-\lambda_j(z-\frac{1}{2})^2} \sum_{r=m}^{i-1} \phi_r(z,t) \left[ b_{r,m} \left( e^{\frac{(z-\frac{1}{2})r}{v_1}} \right) \right] \right\}$$

$$+ \sum_{j=1}^{i-1} \sum_{m=j}^{i} A_{i,j} \sum_{r=m}^{i-1} e^{-\lambda_j(z-\frac{1}{2})^2} \sum_{r=m}^{i-1} g_{r,m} \left[ e^{\frac{(z-\frac{1}{2})r}{v_1}} \phi_r(z,t) \right]$$

$$+ \sum_{j=1}^{i-1} \frac{r_0}{l_\rho} e^{-\lambda_j(z-\frac{1}{2})^2} \sum_{m=j}^{i} \phi_m(t-\frac{1}{2}) + \frac{r_0}{l_\rho} \sum_{\alpha=1}^{i-1} A_{i,\alpha} \sum_{\beta=\alpha}^{i} \frac{1}{b_{\beta,\alpha}}$$

$$\left( e^{-\lambda_\beta(z-\frac{1}{2})^2} \sum_{\omega=\alpha}^{i} d_{\omega,\beta} \phi_\omega(t) \phi_\beta(t) \right)$$

(6.23)
\( l_{r,m}(1z,t') \odot \phi_j(t') \) means that one must first perform the convolution product using \( t' \), then set \( t' = t-(z-1z)/2v_i \).

6.4.2 Step release

\( 1N_i^{S}(z,t) \) is given by (5.13). Using \( \phi_i(t) = B_i(t) \) in Eq. (6.23) we get

\[
2\lambda_i^{S}(z,t) = \sum_{q=1}^{i-1} B_{iq} 2\alpha_i^{q}(z,t) 2\beta_i^{q}(z,t,\lambda_q) + \sum_{j=1}^{i-1} \sum_{m=j}^{i-1} \sum_{r \neq m}^{j} B_{jr} 1\gamma_i^{j,q} 2\alpha_i^{m}(z,t)
\]

\[
= \left[ 2p_i^{m}(z,t,\lambda_q) - 2p_i^{m}(z,t,\Delta_{r,m}) \right] + \sum_{j=1}^{i-1} \sum_{m=j}^{i-1} \sum_{r \neq m}^{j} B_{jr} \frac{1}{2} \gamma_i^{j,q} 2\beta_i^{m}(z,t)
\]

\[
= \sum_{j=1}^{i-1} \sum_{m=j}^{i-1} \sum_{r \neq m}^{j} B_{aq} 1\gamma_i^{j,q} 2\beta_i^{m}(z,t) \theta_j(z,t)
\]

\[
= \left[ 2p_i^{m}(z,t,\lambda_q) - 2p_i^{m}(z,t,\Delta_{r,m}) \right] + \left( \frac{1}{2} \gamma_i^{j,q} \Delta_{r,m} \right) \left[ 2p_i^{m}(z,t,\Delta_{r,m}) - 2p_i^{m}(z,t,\Delta_{r,m}) \right]
\]

(6.24)

where

\[
2p_i^{j}(z,t,\lambda_q) = e^{-\lambda q} \theta_j(z,t)
\]

(6.25)

\[
2p_i^{j}(z,t,\Delta_{mr}) = e^{-\lambda \Delta_{r,m}} \theta_j(z,t)
\]

(6.26)

\[
2G_{\rho}^{j}(z,t) = \frac{1}{1} e^{-\lambda (z-1z)/2v_i} e^{-\lambda z/2v_j} h(\theta_j(z,t))
\]

(6.27)
\[ \ell_{r,m}^{i,j,q} = A_i^{(j)} \ell_{r,m}^{i,j} \ell_{r,m}^{j,q} (\Delta m \rightarrow q) \]  \hspace{1cm} (6.28)

\[ \Theta_{p,j} (z,t) \equiv t - \frac{z^{-1}z}{2v_p} - \frac{z^{-1}z}{1v_j} \]  \hspace{1cm} (6.29)

\( B_{i,q} \) are the coefficients in the Bateman equation: See Chapter 3.

6.4.3 Step-release solution in a two-medium system for a three-member chain, no dispersion

By substituting \( i = 1,2,3 \) into (5.13) and (6.24) we obtain:

First member:

\[ 1^s_{N_1}(z,t) = 1^s_{G_1}(z,t) 1^s_{F_1}(z,t,\lambda_1) B_{11} ; \quad 0 \leq z \leq \frac{1}{z} \]  \hspace{1cm} (6.30)

\[ 2^s_{N_1}(z,t) = 2^s_{G_1}(z,t) 2^s_{F_1}(z,t,\lambda_1) B_{11} ; \quad z \geq \frac{1}{z} \]  \hspace{1cm} (6.31)

Second member:

\[ 1^s_{N_2}(z,t) = 1^s_{A_2}(z,t) ; \quad 0 \leq z \leq \frac{1}{z} \]  \hspace{1cm} (6.32)

\[ 2^s_{N_2}(z,t) = 2^s_{A_2}(z,t) + 2^s_{B_2}(z,t) ; \quad z \geq \frac{1}{z} \]  \hspace{1cm} (6.33)

Third member:

\[ 1^s_{N_3}(z,t) = 1^s_{A_3}(z,t) ; \quad 0 \leq z \leq \frac{1}{z} \]  \hspace{1cm} (6.34)

\[ 2^s_{N_3}(z,t) = 2^s_{A_3}(z,t) + 2^s_{B_3}(z,t) + 2^s_{C_3}(z,t) ; \quad z \geq \frac{1}{z} \]  \hspace{1cm} (6.35)
where:

\[
A_2^s(z,t) = G_2^s(z,t) \left[ B_{21} F_2^1(z,t,\lambda_1) + B_{22} F_2^2(z,t,\lambda_2) \right] \\
+ H_7 B_{11} G_2^1(z,t) \left[ F_2^1(z,t,\lambda_1) - F_2^1(z,t,\lambda_2) \right] \\
+ H_7 B_{11} G_2^2(z,t) \left[ F_2^2(z,t,\lambda_1) - F_2^2(z,t,\lambda_2) \right]
\]

(6.36)

\[
A_3^s(z,t) = G_3^s(z,t) \left[ B_{31} F_3^1(z,t,\lambda_1) + B_{32} F_3^2(z,t,\lambda_2) + B_{33} F_3^3(z,t,\lambda_3) \right] \\
+ H_2 B_{11} G_3^1(z,t) \left[ F_3^1(z,t,\lambda_1) - F_3^1(z,t,\lambda_2) \right] \\
+ H_3 B_{11} G_3^2(z,t) \left[ F_3^2(z,t,\lambda_1) - F_3^2(z,t,\lambda_2) \right] \\
+ H_2 B_{11} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_4 B_{11} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_3 B_{11} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_4 B_{11} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_3 B_{21} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_6 B_{22} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_5 B_{21} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_1) - F_3^3(z,t,\lambda_2) \right] \\
+ H_6 B_{22} G_3^3(z,t) \left[ F_3^3(z,t,\lambda_2) - F_3^3(z,t,\lambda_2) \right]
\]

(6.37)
\[ \begin{align*}
2 \mathcal{B}^s_2(z, t) &\equiv 2H_2 B_{11} G_1^1(z, t) \left[ 2F_1^1(z, t, \Delta_{21}) - 2F_1^1(z, t, \Delta_{11}) \right] \\
&+ 2H_2 B_{11} G_2^1(z, t) \left[ 2F_2^1(z, t, \Delta_{21}) - 2F_2^1(z, t, \Delta_{21}) \right] \\
\quad &+ 2H_3 B_{11} G_1^1(z, t) \left[ 2F_1^1(z, t, \Delta_{31}) - 2F_1^1(z, t, \Delta_{11}) \right] \\
&+ 2H_2 B_{11} G_2^1(z, t) \left[ 2F_2^1(z, t, \Delta_{21}) - 2F_2^1(z, t, \Delta_{21}) \right] \\
&+ 2H_4 B_{11} G_2^1(z, t) \left[ 2F_2^1(z, t, \Delta_{32}) - 2F_2^1(z, t, \Delta_{32}) \right] \\
&+ 2H_3 B_{11} G_3^1(z, t) \left[ 2F_3^1(z, t, \Delta_{31}) - 2F_3^1(z, t, \Delta_{31}) \right] \\
&+ 2H_4 B_{11} G_3^1(z, t) \left[ 2F_3^1(z, t, \Delta_{31}) - 2F_3^1(z, t, \Delta_{31}) \right] \\
&+ 2H_5 B_{21} G_2^2(z, t) \left[ 2F_2^2(z, t, \Delta_{32}) - 2F_2^2(z, t, \Delta_{32}) \right] \\
&+ 2H_5 B_{21} G_3^2(z, t) \left[ 2F_3^2(z, t, \Delta_{32}) - 2F_3^2(z, t, \Delta_{32}) \right] \\
&+ 2H_6 B_{22} G_3^2(z, t) \left[ 2F_3^2(z, t, \Delta_{32}) - 2F_3^2(z, t, \Delta_{32}) \right] \\
\quad &+ 2H_6 B_{22} G_3^2(z, t) \left[ 2F_3^2(z, t, \Delta_{32}) - 2F_3^2(z, t, \Delta_{32}) \right] \\
\end{align*} \] 

(6.38)
\[ 2 \mathcal{C}_3(z,t) = 2H_5^1 H_7 B_{11}^1 \mathcal{G}_2(z,t) \left[ M_{32}^1(z,t,\lambda_1^1) + M_{32}^1(z,t,\lambda_2^1) \right] \\
+ 2H_5^1 H_7 B_{11}^2 \mathcal{G}_2(z,t) \left[ M_{23}^2(z,t,\lambda_1^2) + M_{23}^2(z,t,\lambda_2^2) \right] \\
+ 2H_5^1 H_7 B_{11}^2 \mathcal{G}_2(z,t) \left[ M_{32}^2(z,t,\lambda_1^2) + M_{32}^2(z,t,\lambda_2^2) \right] \\
+ 2H_5^1 H_7 B_{11}^2 \mathcal{G}_2(z,t) \left[ M_{23}^2(z,t,\lambda_1^2) + M_{23}^2(z,t,\lambda_2^2) \right] \]

(6.40)

and where:

\[ \Delta_{r,m} = \frac{\lambda_{m}/v_{m} - \lambda_{r}/v_{r}}{1/v_{m} - 1/v_{r}} \]

(6.41)

\[ 1F_1^j(z,t,\lambda_K) = 1F_2^j(z,t,\lambda_K) = 1F_3^j(z,t,\lambda_K) = e^{-\lambda_K(z/v_j - t)} \]

(6.42)

\[ 1F_1^j(z,t,\lambda_{r,m}) = 1F_2^j(z,t,\lambda_{r,m}) = 1F_3^j(z,t,\lambda_{r,m}) = e^{\Delta_{r,m}(z/v_j - t)} \]

(6.43)

\[ 2F_p^j(z,t,\lambda_K) = e^{-\lambda_K \Theta_j p(z,t)} \]

(6.44)

\[ 2F_p^j(z,t,\lambda_{r,m}) = e^{-\lambda_{r,m} \Theta_j p(z,t)} \]

(6.45)

\[ 1G_1^j(z,t) = 1G_2^j(z,t) = 1G_3^j(z,t) = e^{-z \lambda_j/\sqrt{v_j}} h(t - z/v_j) \]

(6.46)

\[ 2G_p^j(z,t) = e^{-\lambda_p(z - 1) / 2 \Theta_j p} \cdot e^{-\lambda_j z^{1/\sqrt{v_j}} h(\Theta_j p z, t, \frac{z}{1 \rho})} \]

(6.47)
\( h(t) = \text{step function} \)

\[
\ell_{H_1} = \lambda_1 \lambda_2 \ell v_1 \ell v_3 / \left[ (\ell v_1 - \ell v_2)(\ell v_1 - \ell v_3)(\ell \Delta_{21} - \ell \Delta_{31}) \right]
\] (6.48)

\[
\ell_{H_2} = \ell_{H_1} / (\ell \Delta_{21} - \lambda_1)
\] (6.49)

\[
\ell_{H_3} = \ell_{H_1} / (\lambda_1 - \ell \Delta_{31})
\] (6.50)

\[
\ell_{H_4} = \ell_{H_1} / (\ell \Delta_{32} - \lambda_1)
\] (6.51)

\[
\ell_{H_5} = \lambda_2 \ell v_3 / \left[ (\ell v_2 - \ell v_3)(\lambda_1 - \ell \Delta_{32}) \right]
\] (6.52)

\[
\ell_{H_6} = \lambda_2 \ell v_3 / \left[ (\ell v_2 - \ell v_3)(\lambda_2 - \ell \Delta_{32}) \right]
\] (6.53)

\[
\ell_{H_7} = \lambda_1 \ell v_2 / \left[ (\ell v_1 - \ell v_2)(\lambda_1 - \ell \Delta_{21}) \right]
\] (6.54)

\[
M^\beta_{rm}(z,t,\lambda_1) = 2F_m(z,t,\lambda_1) - 2F_m(z,t,2\Delta_{rm})
\] (6.55)

\[
M^\beta_{rm}(z,t,1\Delta_{21}) = \frac{(2\Delta_{rm} - \lambda_1)}{\left(2\Delta_{rm} - 1\Delta_{21} \right)} \times \left[ 2F_m(z,t,2\Delta_{rm}) - 2F_m(z,t,1\Delta_{21}) \right]
\] (6.56)

\[
\Theta_{jp}(z,t) = t - \frac{z-1}{2v} - \frac{1}{v_j}
\] (6.57)

The application of these equations to a numerical problem will be demonstrated in a later report.
6.5 Recursive Formula for a Two-Medium System with Dispersion

For this case we use the transport equation with the source term. We have the following situation as shown in Figure 6.2.

![Diagram of two-medium system with dispersion](image)

Figure 6.2. Two-medium system with dispersion.

The interface between the two-medium is set at \( z = 0 \). The repository is set in the first medium, at \( z_1 \).

The boundary condition at the media boundary has been discussed in Chapter 6.2. We have therefore the problem to solve:

\[
I_i^1 \frac{d}{dt} N_i(z, t) = \lambda_i - 1 \frac{d}{dz} \frac{1}{K_i - 1} N_i - 1 + \Phi_i(t) \delta(z - z_1) \quad ; \quad z \leq 0
\]

(6.58)

where \( \Phi_i(t) \) has dimensions of \( (m^2 \cdot yr)^{-1} \)

\[
2 \frac{d}{dt} N_i(z, t) = \lambda_i - 1 \frac{d}{dz} N_i - 1 \quad ; \quad z \geq 0
\]

(6.59)

with

I.C.

\[
N_i(z, 0) = 2 N_i(z, 0) = 0
\]

(6.60)

B.C.

\[
N_i(-\infty, t) = \text{bound}
\]

(6.61)

\[
N_i(+\infty, t) = \text{bound}
\]

(6.62)
\[ \begin{align*}
N_1(0, t) &= 2N_1(0, t) \quad (6.63) \\
\frac{1}{2} \frac{d_i}{dz} \frac{1}{\frac{d_i}{dz}} &= \frac{\partial N_i}{\partial z} \quad \text{at } z = 0 \quad (6.64)
\end{align*} \]

This problem is solved by the Green's method. We seek first the solutions for a unit pulse (in time and space) in the first medium, then in the second. We solve

\[ \begin{align*}
\frac{1}{L_1} \n_1(z, t; \xi, \tau) &= \delta( z - \xi) \delta( t - \tau) \quad ; \quad \xi < 0, \ z \leq 0, \ 0 \leq \tau \leq t \\
\frac{2}{L_1} \m_1(z, t; \xi, \zeta) &= 0 \quad ; \quad \xi < 0, \ z \geq 0, \ 0 \leq \tau \leq t
\end{align*} \quad (6.65) \]

with the initial conditions (6.60) and the boundary conditions (6.61) to (6.64).

Then we solve

\[ \begin{align*}
\frac{1}{L_1} \n_1(z, t; \xi, \tau) &= 0 \quad ; \quad \xi > 0, \ z \leq 0, \ 0 \leq \tau \leq t \\
\frac{2}{L_1} \m_1(z, t; \xi, \tau) &= \delta(z - \xi) \delta( t - \tau) \quad ; \quad \xi > 0, \ z \geq 0, \ 0 \leq \tau \leq t
\end{align*} \quad (6.66) \]

with the same initial and boundary conditions.

\[ L_1 n_1(z, t; \xi, \tau) \] represents the response in the first medium to a pulse injected into the first (left) medium at \( z = \xi \) and \( t = \tau \).

\[ L_1 m_1(z, t; \xi, \tau) \] represents the response in the second medium to a pulse injected into the first medium at \( z = \xi \) and \( t = \tau \). In the same way, \( R n_1 \) and \( R m_1 \) represent, respectively, the responses in the first
or second medium to a pulse injected into the second (right-hand) medium. Equations (6.65) and (6.66) are solved by taking the Laplace transform with respect to \( t \). The equations then become ordinary differential equations of second order in one variable \( z \). These can be solved, and they yield the Laplace transformed solutions \( L_{n_1}^-(z,s) \), \( L_{m_1}^-(z,s) \), \( R_{n_1}^-(z,s) \), \( R_{m_1}^-(z,s) \). By the use of Laplace transform tables, one finds \( L_{n_1}^+(z,t) \), \( L_{m_1}^+(z,t) \) etc. These functions: \( L_{n_1}^+ \), \( L_{m_1}^+ \), \( R_{n_1}^- \), \( R_{m_1}^- \), play then the role of a Green kernel. The final result is then:

\[
i_{n_1}^+(z,t) = R_{n_1}^-(z,t) + L_{n_1}^+(z,t) \quad ; \quad z < 0
\]

\[
2i_{n_1}^+(z,t) = R_{m_1}^+(z,t) + L_{m_1}^+(z,t) \quad ; \quad z > 0
\]

\[
R_{n_1}^+(z,t) \equiv \int_0^t \left[ \int_0^\infty R_{n_1}(z,t;\xi,\tau)^2 f_{i_1}(\xi,\tau)d\xi \right] d\tau \quad ; \quad z < 0
\]

\[
L_{n_1}^+(z,t) \equiv \int_0^t \left[ \int_0^\infty L_{n_1}(z,t;\xi,\tau) f_{i_1}(\xi,\tau)d\xi \right] d\tau \quad ; \quad z < 0
\]

\[
R_{m_1}^+(z,t) \equiv \int_0^t \left[ \int_0^\infty R_{m_1}(z,t;\xi,\tau)^2 f_{i_1}(\xi,\tau)d\xi \right] d\tau ; \quad z > 0
\]
\[ L_{m_1}(z, t) \equiv \int_0^t \left[ \int_{-\infty}^0 L_{m_1}(z, t; \xi, \tau) \frac{1}{f_1(\xi, \tau)} \, d\xi \right] \, d\tau; \quad z > 0 \]  

\[ R_{m_1}(z, t; \xi, \tau) = \frac{1}{2^{\gamma_1}} e^{(z-\xi)/2} \cdot \begin{cases} 
\frac{1}{G_+} 2 \frac{1}{p_4(\xi, z-z)} \frac{1}{p_4(-\xi, z-\tau)} - \frac{1}{2G_-} \frac{1}{p_5(z-\xi, \xi, z-\tau)} & ; \xi > 0 \\
\frac{1}{2^{\gamma_1}} \frac{1}{p_4(z-\xi, \xi, z-\tau)} - \frac{2^{\gamma_1}}{G_-} \frac{1}{p_3(z+\xi, \xi, z-\tau)} & ; \xi < 0 
\end{cases} \]  

\[ \begin{align*}
R_{m_1}(z, t; \xi, \tau) &= \frac{1}{2^{\gamma_1}} e^{(z-\xi)/2} h(t-\tau) \begin{cases} 
2 \frac{1}{p_4(z-\xi, t-\tau) h(z-\xi)} + \\
2 \frac{1}{p_4(z-\xi, t-\tau) h(z-\xi)} + & 2 \frac{1}{p_4(z+\xi, t-\tau)} + \\
+ \frac{2^{\gamma_1}}{G_-} \frac{1}{p_3(z+\xi, t-\tau)} & ; \xi > 0 
\end{cases} \end{align*} \]
\( L_{m_1}(z, t, \xi, \tau) = \frac{e^{z/2t}}{G} \cdot e^{-\xi/2t} \cdot h(t-\tau) \left\{ 1, 2p^4_4(-\xi, z, t-\tau) \right\} - \\
- 1y_1 \frac{1, 2p^4_5(\xi, z, t-\tau)}{} \) \quad ; \quad \xi < 0 \tag{6.76}

\( 1f_{i_1}(\xi, \tau) = \frac{\lambda_{i-1}}{\lambda_{i-1}} \cdot \frac{1}{v_{i-1}} \cdot \mathcal{N}_{i-1}(\xi, \tau) + \frac{\phi_i(\tau)}{v} \cdot \delta(\xi - 1z) \) \quad ; \quad \xi < 0 \tag{6.77}

\( 2f_{i_1}(\xi, \tau) = \frac{\lambda_{i-1}}{\lambda_{i-1}} \cdot \frac{2}{v_{i-1}} \cdot \mathcal{N}_{i-1}(\xi, \tau) \) \quad ; \quad \xi > 0 \tag{6.78}

\( l_{p^4_1}(z, t) = \frac{1}{\sqrt{\pi t}} \cdot e^{-[\xi^2/4t] \cdot v_{i_1} \cdot d_1} \) \quad ; \quad t > 0 \tag{6.79}

\( l_{p^4_2}(z+\xi, t) = \int_{0}^{t} e^{-\lambda_{i}u} \cdot l_{p^4_1}(z+\xi, t-u) du \) \quad ; \quad t > 0 \tag{6.80}

\( \xi_{p^4_2}(z, t) = \int_{\xi}^{t} \frac{1}{2(t-u) \cdot v_{i_1} \cdot d_1} \cdot l_{p^4_1}(z, t-u) \cdot \left\{ \frac{v_{i_1}^{1/2} \cdot t_{i_1}^{1/2} \cdot u}{\sqrt{\pi u}} + \frac{v_{i_1}^{1/2}}{\sqrt{\pi u}} \cdot e^{-\lambda_{i}u} \cdot \text{erf}\left(\frac{1}{2} \cdot \sqrt{\frac{v_{i_1}^{1/2}}{\lambda_{i}}} \cdot u\right) \right\} \) \quad ; \quad t > 0 \tag{6.81}

\( \xi_{p^4_4}(x, y, t) = \int_{0}^{t} \frac{e_{p_4}^4(x, t-u)}{2 \cdot \sqrt{v_{i_1} \cdot d}} \cdot \left[ \frac{1}{2} \cdot K_{i_1}(y, u) + K_{y_i} \cdot K_{p_1}(y, u) \right] \) \quad ; \quad t > 0 \tag{6.82}
\[ \ell \gamma_i \equiv \sqrt{\frac{\ell d_i}{\ell v_i}} \]  

(6.88)

\[ \ell \omega_i \equiv \lambda_i + \frac{\ell v_i}{4 \ell d_i} \]  

(6.89)
\begin{align*}
\kappa_i^{(x,y,t)} &= \int_0^t \left[ \frac{x^2}{2(t-u)K_v^d} - 1 \right] \kappa_i^{(x,t-u)} \kappa_\delta^{(y,u)} \, du \quad ; \quad t > 0 \quad (6.83) \\
\kappa_i^{(z,t)} &= \frac{e^{-\lambda_i t}}{2} \left\{ e^{-z/2K_v^d} \text{erfc} \left[ \frac{z}{2\sqrt{tK_v^dK_v^d}} - \frac{1}{2} \sqrt{\frac{K_v^d}{t K_v^dK_v^d}} \right] \right. \\
& \quad \left. + e^{z/2K_v^d} \text{erfc} \left[ \frac{z}{2\sqrt{tK_v^dK_v^d}} + \frac{1}{2} \sqrt{\frac{K_v^d}{t K_v^dK_v^d}} \right] \right\} \quad ; \quad t > 0 \quad (6.84) \\
\kappa_i^{(z,t)} &= \frac{e^{-\lambda_i t}}{2} \left\{ e^{-z/2K_v^d} \text{erfc} \left[ \frac{z}{2\sqrt{tK_v^dK_v^d}} - \frac{1}{2} \sqrt{\frac{K_v^d}{t K_v^dK_v^d}} \right] \right. \\
& \quad \left. - e^{z/2K_v^d} \text{erfc} \left[ \frac{z}{2\sqrt{tK_v^dK_v^d}} + \frac{1}{2} \sqrt{\frac{K_v^d}{t K_v^dK_v^d}} \right] \right\} \quad ; \quad t > 0 \quad (6.85) \\

\text{Notice: The superscript } i \text{ in } \kappa_i^{(x,y,t)}, \kappa_i^{(z,t)} \text{ means that one must use } \kappa_v, \kappa_d, \kappa_i \text{ etc. whenever } \kappa_v, \kappa_d, \kappa_i \text{ etc. appear in the definition.} \\
G_- &= \frac{2d^1_{v-1d^2_{v}}}{1^v_2^v} \quad (6.87) \\
G_+ &= \frac{2d^1_{v-1d^2_{v}}}{1^v_2^v} \quad (6.88)
\end{align*}
7. CONCLUSIONS

The research in FY-1979 is aimed to elucidate the following problems:

1) The fundamental transport equation governing nuclide migration,
2) The relation among several modes of nuclide release from the waste,
3) Recursive and general solutions of an i-member chain migration for one-dimensional transport and for arbitrary release modes,
4) Application of the recursive and the general solutions to a three-member decay chain and the elucidation of the migration features for some of the important multi-member decay chains,
5) Nuclide migration through multi-layer media.

To summarize, the following results have been obtained:

1. The fundamental transport equations governing the migration of nuclide decay chain were derived, based on a phenomenological approach and with the help of the volume-average concept. The equations were reduced to one-dimensional transport, with local chemical equilibrium, as considered in this report.

2. The relationship among several release modes has been discussed. The release modes include a) impulse, b) step, c) band, d) linear leach rate, e) fractional leach rate characteristic of an individual nuclide. The transformation to yield different solutions from the solution of a representative release mode has been discussed.
3. A general superposition theorem has been developed for analyzing the propagation of chromatographic bands with radioactive decay. This provides an exact method for constructing the space-time dependent concentration resulting from a source of finite duration (band release) by superposing time-displaced solutions for a source of infinite duration (step release).

4. Recursive solutions of the one-dimensional transport equations with and without dispersion have been developed, both for a generalized source boundary condition and for a generalized concentration boundary condition. An explicit (general) solution for any radionuclide in an i-member decay chain in one dimension and without dispersion has been developed.

5. Explicit solutions for three-member chains in one-dimensional transport have been developed for a plane source of constant leach rate, with and without dispersion, for a source boundary condition and for the specification of time-dependent concentration of dissolved radionuclides at the source. The solution has been programmed.

Explicit solutions for an i-member decay chain without dispersion have been developed for the above five release modes.

The migration features for the most important two decay chains, $^{234}\text{Y} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ and $^{237}\text{NP} \rightarrow ^{233}\text{Pa} \rightarrow ^{233}\text{U} \rightarrow ^{229}\text{Th} \rightarrow ^{225}\text{Ra}$, have been elucidated with the help of the above solutions. The isopleths for $^{226}\text{Ra}$ in the water pathway have been demonstrated, in which the peak concentration of a nuclide in the moving sorption band can increase with distance travelled through the medium.
parametric analysis for the migration features has been carried out for the dispersion coefficient, the leaching time, the sorption retardation constant and the amounts of decay daughters in the waste at the beginning of waste dissolution.

6. An analytical expression for the one-dimensional migration of nuclides through multi-layered media of different hydrological and sorptive properties has been developed. This expression is of the recursive type with respect to media and nuclide decay. An analytical explicit solution for two media and a three-member decay chain has been obtained for the dispersion free case. The recursive solution for two media with dispersion has been developed.
8. NOMENCLATURE

$A$ : Interfacial area in the averaging domain

$A_{i}$ : Coefficient defined by (2.55)

$A_{i}(j) = \prod_{r=j}^{i-1} \left( \frac{\lambda_{r}}{v_{r}} \right)$ : Defined by (4.113)

$\mathcal{A}_{i}$ : Function defined by (6.36) and (6.37)

$a$ : Interfacial area per unit volume of geological media

$B_{i}$ : Coefficient defined by (2.55)

$B_{i}(t)$ : Bateman equation

$B_{ij}$ : Bateman coefficient defined by (3.12)

$B'_{ij}$ : Modified Bateman coefficient defined by (3.23)

$B_{m} = B_{m}^{(ij)} = \prod_{r=j}^{i} \Gamma_{rm}$ : Defined by (4.121)

$\mathcal{B}_{i}$ : Function defined by (6.38) and (6.39)

$C_{k\alpha}$ : Concentration of the $k^{th}$ chemical species in the $\alpha$ phase

$\dot{C}_{k\alpha}$ : Fluctuating-concentration of the $k^{th}$ chemical species in the $\alpha$-phase

$C_{m}(s) = \left[ \prod_{r=j}^{i} \left( \mu_{r} - \mu_{m} \right) \right]^{-1}$ : Defined by (4.116)

$\mathcal{C}_{3}$ : Function defined by (6.40)

$D$ : Axial effective dispersion coefficient (mean value)

$D^{*}$ : Axial dispersion coefficient defined by (4.41)
\[ D_i \] : Effective dispersion coefficient for the \( i^{th} \) nuclide

\[ D_f \] : Dispersion coefficient in the fluid phase

\[ D_{\text{df}} = D_{d\text{f}} + D_{m\text{f}} \] : Dispersion coefficient tensor

\[ D_s \] : Molecular diffusion coefficient in the solid phase

\[ D_{\text{df}} \] : Fluid dispersion coefficient tensor in the fluid phase

\[ D_{\text{mk}} \] : Molecular diffusion coefficient of the \( k^{th} \) species in the \( \alpha \)-phase

\[ D_{\text{mf}} \] : Molecular diffusion coefficient in the fluid phase

\[ D_{\text{ij}} \rightarrow D_{\text{rm}} \] : Defined by (4.122)

\[ \mathcal{L}_{D_{ii}} = \frac{\mathcal{L}_{D_i}}{\mathcal{L}_v} \] : Defined by (6.4)

\[ E(i,j,k) \] : Function defined by (5.33) or (5.40)

\[ F(\alpha, \beta) \] : Gaussian distribution function defined by (4.33)

\[ f_{i\alpha}(x,t) \] : Source term of the \( i^{th} \) nuclide in the \( \alpha \)-phase

\[ f_{i}(z,t) \] : Source term of the \( i^{th} \) nuclide in the water phase

\[ f_{i}(t) \] : \( \phi_i(t)\delta(z) \) : Defined by (4.23)

\[ \mathcal{L}_{f_i} \] : Function defined by (6.77) and (6.78)

\[ \mathcal{L}_{f_j} \] : Function defined by (6.42) to (6.45) or (6.25) and (6.26)

\[ G_i(z,t; \xi, \tau), G_i(z; \xi, \tau) \] : Green's function

\[ G_+, G_- \] : Function defined by (6.87) and (6.86)

\[ \mathcal{L}_{G_j} \] : Function defined by (6.46) and (6.47) or (6.27)

\[ g_i(t) \] : Function defined by (4.47b)

\[ g_{\text{rm}}(z,t) \equiv g_{\text{rm}}(t) \] : Function defined by (4.124)
\[ H_i(z_o, t) \]: Ingestion toxicity of \( i^{th} \) nuclide (cumulative)

\[ \ell \]

\[ l_{ijq} \]: Function defined by (6.48) to (6.54)

\[ H_{rm} \]: Function defined by (6.28)

\[ H(\xi \in \chi) \]: Function defined by (2.5)

\[ h(t-T) \]: Heaviside step function

\[ \ell_I = \ell + 1 \rho / \ell \rho \]

\[ j_{k\alpha} \]: Relative flux of the \( k^{th} \) chemical species in \( \alpha \)-phase

\[ K_i \]: Overall sorption retardation coefficient defined by (2.53)

\[ K_{D,i} \]: Overall distribution coefficient defined by (2.57)

\[ k \]: Leaching rate constant

\[ k_i \]: Leaching rate constant for the \( i^{th} \) nuclide species defined by (3.18)

\[ k_{Ak} \]: Reaction rate constant for the \( k^{th} \) chemical species defined by (2.58)

\[ K_{D,k} \]: Distribution coefficient of the \( k^{th} \) chemical species between solid and water phase (2.49)

\[ K_{D,i} \]: Distribution coefficient of the \( i^{th} \) nuclide between solid and water phases

\[ k_l \]: Rate coefficient for mineralization reaction

\[ k_{Mk} \]: Overall mass transfer coefficient for the \( k^{th} \) chemical species

\[ k_{R,k\alpha} \]: Formation constant of the \( t^{th} \) chemical species from the \( k^{th} \) species in the \( \alpha \)-phase
L : Length of repository zone

\( \hat{L}_i, \tilde{L}_i \) : Partial differential operators defined by (3.34) and (6.2)

l : Unit tensor

\( M_T \) : Total amount of waste material

\( M_i(t) \) : Amount of the \( i^{th} \) nuclide in the repository at time \( t \)

\( M_i^0 \) : Initial amount of the \( i^{th} \) nuclide in the repository

\( M_{\beta \tau m} \) : Function defined by (6.55) and (6.56)

\( \omega_{\alpha, i}(z,t; \xi, \tau) \) : Green's function satisfying (6.65) and (6.66) for \( \alpha = R \) or \( L \)

\( \mathcal{N}_{(i)}(z,t; \Phi_i(t)) \) : function defined by (6.19)

\( N_i^0 \) : Initial nuclide concentration in the water phase

\( N_i(t) \) : Concentration of the \( i^{th} \) nuclide in water phase at the exit of the repository

\( N_{\alpha i}(z,t) \) : Concentration of the \( i^{th} \) nuclide in the \( \alpha \)-phase

\( N_i(z,t) \) : Concentration of the \( i^{th} \) nuclide in the water phase

\( N_i(j)(z,t) \) : Concentration of the \( i^{th} \) nuclide due to the \( j^{th} \) nuclide released from the waste source

\( N_i(z,t)^* \) : Step release solution for the \( i^{th} \) nuclide concentration, satisfying the boundary condition (3.40)

\( N_i,r(t) \) : Concentration of the \( i^{th} \) nuclide defined by (4.84)

\( N_{i,r}(t) \) : Step release solution for the \( i^{th} \) nuclide concentration, satisfying the boundary condition (3.40)

\( n_{\alpha} \) : Unit normal vector pointing out of the interface from the \( \alpha \)-phase

\( n_i(t) \) : Amount of nuclide \( i \) per unit amount of waste
\( n_i(p,t) \) : Laplace transform of \( N_i(z,t) \) with respect to \( z \)

\( n_i^{(j)}(p,t) \) : Laplace transform of \( N_i^{(j)}(z,t) \) with respect to \( z \)

\( \alpha n_i(z,t;\xi,\tau) \) : Green's function satisfying (6.65) and (6.66)

\( \alpha = R \) or \( L \)

\( \ell_k^i p_j \) : Function defined by (6.81) and (6.82)

\( \ell_p^i p_j \) : Function defined by (6.79) and (6.80), (6.84) and (6.85)

\( p \) : Variable for Laplace transform with respect to \( z \)

\( Q \) : Volumetric flow rate of water

\( (RCG)_i \) : Radioactivity Concentration Guide from 10 CFR 20 Appendix B

\( R_{k\alpha} \) : Birth rate of the \( k^{th} \) chemical species in the \( \alpha \)-phase

\( r_{i-1,i}^{(z,t)} \) : Inverse transform of \( r_{i-1,i}(p,t) \)

\( r_{i-1,i}(p,t) \) : Function defined by (4.55b)

\( S \) : Effective cross-sectional area for flow of the water phase

\( S_{\alpha} \) : Cross-sectional area of \( \alpha \)-phase

\( S_{i-1}(z,t) \) : Function defined by (4.45b)

\( S_k^i \) : Number of atoms per molecule of the \( i^{th} \) nuclide existing in the \( k^{th} \) chemical form

\( s \) : Variable for Laplace transform with respect to time \( t \)

\( s_{i-1}(p,t) \) : Laplace transform of \( S_{i-1}(z,t) \)

\( S_{i-1}^{i-1} \) : Fraction of the decay of the \((i-1)\)th nuclide which forms the \( i \) \( 1 \) chemical form

\( t \) : Time

\( T \) : Leach time, i.e., time for complete dissolution of the radioactive waste
T(p,t) : Transform kernel defined by (4.50)
U₁(z,t) : Function defined by (4.44)
u₁(p,t) : Laplace transform of U₁(z,t)
V : Volume of averaging domain of water phase volume
v : water phase velocity
va : Velocity of averaging domain or water phase volume
v_i : v/K_i
v_f : Fluid phase velocity
w : Interface velocity
X : Position vector measured from the reference coordinate
x : Position vector relative to X
x_b : Position vector at interface
z : Distance along water pathway
z_l : Repository position
z_l : Length of the i-th layer
z_1 : Relative distance from the (l-1)-layer boundary
α_i = λ_i k_i : Defined by (4.42)
β_i = -λ_i γ i_1 : Defined by (4.42)
β_ij : Parameter defined by (5.32)
γ = v/2D : Defined by (4.42)
\( e_i = \sqrt{\frac{\xi_i}{v_i}} \) : Defined by (6.88)
γ_i = Parameter defined by (5.32)
Γ_r_m = v_r-1 - v_m-1
Δ_i_r = (λ_i/v_i - λ_r/v_r)/(v_i-1 - v_r-1)
δ(t) : Dirac delta function
ε : Porosity
\( \varepsilon_\alpha \): Volume fraction of the \( \alpha \)-phase

\( \Theta_{ip} \): Function defined by (6.29)

\( \eta_i \): \( D_i/K_i \)

\( \kappa \): \( D/v \)

\( \Lambda_i \): \( \lambda_i + k_i \)

\( \Lambda_{kj} \): \( \lambda_k/v_k - \lambda_j/v_j \)

\( \lambda_i \): Decay constant of the \( i^{th} \) nuclide

\( \mu_\lambda \): \( (\delta + \lambda_\lambda)/v_\lambda \) : Defined by (4.110c)

\( \nu_{\lambda-1} \): \( K_{\lambda-1} \lambda_{\lambda-1}/K_{\lambda} \lambda_\lambda \) : Defined by (4.110c)

\( \xi \): Distance

\( \rho \): Density of water phase

\( \rho_\alpha \): Density of the \( \alpha \)-phase

\( \tau \): Time

\( \Phi_{i}(t) \): Function representing functions \( \lambda_{\phi_1}(t), \lambda_{\phi_2}(t), \ldots, \lambda_{\phi_i}(t) \)

\( \Phi_{i}(t) \): Function defined by (6.17) or (6.58)

\( \phi_{i}(t) \): Functions defined by (4.21) or (4.41)

\( \psi \): Dynamical property

\( \psi_\alpha \): Dynamical property in the \( \alpha \)-phase

\( \psi_{\alpha} \): Dynamical property defined by (2.6)

\( \omega_i \): Parameter defined by (6.89)

**Subscript**

\( i \): Nuclide \( i \)

\( k \): Chemical species \( k \)

\( \alpha \): Phase
Superscript

\( b \) : Band release
\( e \) : Exponential release
\( I \) : Impulse release
\( p \) : Preferential release
\( s \) : Step release
\( \ell \) : \( \ell^{th} \) layer (superscript at left-hand side)
\( \sim \) : Volume average defined by (2.8)
\( \wedge \) : Volume average defined by (2.7)
9. REFERENCES


H4 K. Higashi, M. Harada, F. Iwamoto, T. H. Pigford, "Migration Behavior of the $^{234}$U $\rightarrow$ $^{230}$Th $\rightarrow$ $^{226}$Ra $\rightarrow$ Decay Chain", Trans. ANS 33, 386 (1979).


APPENDIX

Appendix A. Computer Code MGRATO3 (UCB-NE-10)

The computer code MGRATO3 (UCB-NE-10) has been developed to calculate the one-dimensional migration of radionuclide chains in a geologic medium for several release modes such as the step, the band, the exponential and the preferential release. Eqs. (5.29), (5.30), and (5.31) are used as solutions for first three members in the case of the step release mode, and the superposition relation (5.35) is then used to obtain the band-release solution. Section A.1.4 presents the three-member solutions used for the exponential and preferential release modes described in Sect. 3.2.2, for the case of longitudinal dispersion.

A.1 Program Description

A.1.1 Overall Structure

MGRATO3 (UCB-NE-10) is written in FORTRAN IV for the CDC-6600/7600 computer, and consists of a main program, five sub-programs and four function programs as shown below.

Main Program: MGRATO3

Sub-programs: CALC : Calculation of $N_i(z,t)$
GAUSS : Numerical Integration
PRINTER: Print the result
TITLE1 : Print the Initial Conditions
TITLE2 : Print the title for the result
Function : EFN : Calculation of \( E(i,j;k) \)
Programs
ERF : \( \text{erf}(x) \) by a rational approximation
YNM : Function for Numerical integration
GMF : Calculation of \( Y_{ijk} \)

The overall structure of MGRAI03 (UCB-NE-10) is shown in Figure A.1, and a program listing is presented in A.1.5.

A.1.2 Input/Output

A.1.2.1 Input

Nine input cards are required to execute the program for a three-member chain. Once these cards are read and executed, one can successively execute the program in the same run only by adding the first card, and cards to change parameters may be added. The end card of the data should be a card numbered 99. By this card, the program identifies the end of data. A schematic explanation of the input data cards is illustrated in Figure A.2. The instructions for data cards are attached in Supplement A.1.
Figure A.1. Overall structure of MGRATO3 (UCB-NE-16)
Figure A.2. Arrangement of data cards for case study.

A.1.2.2 Output

As seen in Figure A.1, MGRAT03 (UCB-NE-10) prints out input data first, and then the results of calculation are printed out for a fixed $z$ or a fixed $t$ specified in the 5th input card. The print of input data contains parameters of geologic system, parameters which characterize the release mode, time, and distance, and which specify the range of calculation and data about radionuclides. A sample of a printed output is illustrated in Supplement A.2.

Results are printed out in two different ways. When the third and fourth columns in the 5th card contain any number other than unity, the following outputs are given:
1) Ratio of the concentration between nuclide \( i \) and the initial concentration of 1st nuclide

\[
\frac{N_i(z,t)}{N_i^0} \quad (A.3)
\]

2) Ratio of radioactivity concentration

\[
\frac{\lambda_i N_i(z,t)}{\lambda N_i^0} \quad (A.4)
\]

When unity is specified as in the third and fourth column of 5th card, two expressions of the discharge rate of nuclide \( i \) are printed out.

1) Radioactivity discharge rate

\[
\frac{\lambda_i N_i(z,t)Q/C_i}{C_i / \text{yr}} \quad (A.5)
\]

2) Ingestion hazard discharge rate

\[
\frac{\lambda_i N_i(z,t)Q/C_i}{(\text{RCG})_i} \quad (\text{m}^3\text{-H}_2\text{O/yr}} \quad (A.6)
\]

where \( C_i : 1.16683 \times 10^{18}, \text{dpy/Ci} \)

\( Q : \text{Flow rate of groundwater, m}^3/\text{yr} \)

\((\text{RCG})_i : \text{Radioactivity Concentration Guide, Ci/m}^3\text{-H}_2\text{O}\)

Moreover, if a fixed \( z \) is taken for calculation of the discharge rate here, the cumulative ingestion toxicity of nuclide \( i \), \( H_i(z_0,t) \) (m\(^3\)-H\(_2\)O) at time \( t \) and at distance \( z_0 \) where the nuclide is discharged to the biosphere in also printed out. The cumulative ingestion toxicity is defined as the total volume of water required to dilute the accumulated discharged nuclide \( i \) to the RCG level for drinking water. \( H_i(z_0,t) \) is given by:
\begin{align*}
H_i(z_o,t) &= \frac{G\lambda_i}{C(RCG)} \sum_{j=1}^{i} \int_{0}^{t} \sum_{m=1}^{j} b_{ij} N(z_o,\theta) e^{-\lambda_j (t-\theta)} d\theta \quad (A.7)
\end{align*}

where

\begin{align*}
b_{ij} &= \left( \frac{1}{\lambda_i} \prod_{r=m}^{i} \lambda_r \right) \left[ \prod_{l=m}^{i} \frac{(\lambda_l - \lambda_j)}{l \neq j} \right] \quad (A.8)
\end{align*}

\(H_i(z_o,t)\) is calculated by a numerical integration of (A.7) in the code.

A.1.3 Calculation of \(E(i,j;k)\)

The solutions to the one-dimensional transport equation are given by linear combinations of \(E(i,j;k)\) functions. In the case of \(\gamma_{ijk} > 0\), \(E(i,j;k)\) consists of the exponential function and the complementary error function as shown in Eq. (5.33b). The calculation of the error function in the code is made by rational approximation shown below:

\begin{align*}
\text{erf}(x) &= 1 - \frac{1}{(1 + a_1 x + a_2 x^2 + \ldots + a_6 x^6)^{1/6}} + \varepsilon(x) \quad (A.1)
\end{align*}

\(|\varepsilon(x)| \leq 3 \times 10^{-7}\)

\begin{align*}
a_1 &= 0.07052 \quad 30784 \\
a_2 &= 0.04228 \quad 20123 \\
a_3 &= 0.00927 \quad 05272 \\
a_4 &= 0.00015 \quad 20143 \\
a_5 &= 0.00027 \quad 55672 \\
a_6 &= 0.00004 \quad 30638
\end{align*}

and

\begin{align*}
\text{erf}(x) &= 1 - (a_1 t + a_2 t^2 + \ldots + a_5 t^5) e^{-x^2} + \varepsilon(x) \quad (A.2)
\end{align*}

\(t = \frac{1}{1 + px}, \quad \varepsilon(x) \leq 1.5 \times 10^{-7}\)
\[ p = 0.32759 \ 11 \quad a_1 = 0.25482 \ 9592 \]
\[ a_2 = -0.28449 \ 6736 \quad a_3 = 1.41141 \ 3741 \]
\[ a_4 = -1.45315 \ 2027 \quad a_5 = 1.06140 \ 5429 \]

Due to the approximation, the absolute error of the result is less than equal to \(3 \times 10^{-7}\).

\(\gamma_{ijk}\), an argument of the complementary error function, sometimes becomes negative when the velocity of the water is small and the effect of the dispersion is important. An earlier code, GETOUT (DI), cannot operate when \(\gamma_{ijk} < 0\). In the present code MGRATO3 (UCB-NF-10), if \(\gamma_{ijk} < 0\), a numerical integration of Eq. (5.33a) is carried out in the subroutine EFN to calculate the \(E(i,j;k)\) function.

A.1.4 Three-member solution for the preferential-release mode

The analytical solutions for the first three members, source term release mode given by the preferential-release mode, are summarized below:

\[ N'_1(x,t) = B'_{11}(E(1,1;1)) \quad (A.9) \]

\[ N'_2(z,t) = B'_{22}E(2,2;2) + B'_{21}E(1,1;2) \]

\[ + \frac{B'_{11} \lambda_1}{\nu_1(\Lambda_{12}-\Lambda_{11} \Gamma_{12})} \left[ E(1,1;2)-E(1,1;1)+E(1,2;1)-E(1,2;2) \right] \quad (A.10) \]
\[ N_3^{\prime}(z,t) = B_{33}^{\prime} E(3,3;3) + B_{32}^{\prime} E(2,2;3) + B_{31}^{\prime} E(1,1;3) \]

\[ + \frac{B_{22}^\prime \lambda_2}{\nu_2 (\Lambda_{23} - \Lambda_1^0 \Gamma_{23}^0)} \left[ E(2,2;3) - E(2,3;3) + E(2,3;2) - E(2,2;2) \right] \]

\[ + \frac{B_{21}^\prime \lambda_2}{\nu_2 (\Lambda_{23} - \Lambda_{11}^1 \Gamma_{23}^1)} \left[ E(1,1;3) - E(2,3;3) + E(2,3;2) - E(1,1;2) \right] \]

\[ + \frac{B_{11}^\prime \lambda_1 \lambda_2 E(1,1;3)}{\nu_1 \nu_2 (\Lambda_{13} - \Lambda_1 \Gamma_{13}) (\Lambda_{23} - \Lambda_1 \Gamma_{23})} + \frac{B_{11}^\prime \lambda_1 \lambda_{23} E(1,3;3)}{\nu_1 \nu_2 (\Lambda_{13} - \Lambda_1 \Gamma_{13})(\Gamma_{23} \Lambda_{13} - \Gamma_{13} \Gamma_{23})} \]

\[ + \frac{B_{11}^\prime \lambda_1 \lambda_{23} E(2,3;3)}{\nu_1 \nu_2 (\Lambda_{23} - \Lambda_1 \Gamma_{23})(\Gamma_{13} \Lambda_{23} - \Gamma_{23} \Lambda_{12})} + \frac{B_{11}^\prime \lambda_1 \lambda_{23} E(1,3;2)}{\nu_1 \nu_2 (\Lambda_{12} \Lambda_{32} \Gamma_{12} \Gamma_{32})(\Lambda_{12} - \Lambda_1 \Gamma_{12})} \]

\[ + \frac{B_{11}^\prime \lambda_1 \lambda_{32} E(2,3;2)}{\nu_1 \nu_2 (\Lambda_{32} - \Lambda_1 \Gamma_{32})(\Gamma_{12} \Lambda_{32} - \Gamma_{32} \Lambda_{12})} + \frac{B_{11}^\prime \lambda_1 \lambda_{32} E(1,2;2)}{\nu_1 \nu_2 (\Lambda_{12} \Lambda_{32} \Gamma_{12} \Gamma_{32})(\Gamma_{32} \Lambda_{12} - \Gamma_{12} \Lambda_{32})} \]

\[ + \frac{B_{11}^\prime \lambda_1 \lambda_{31} E(1,1;1)}{\nu_1 \nu_2 (\Lambda_{31} - \Lambda_1 \Gamma_{31})(\Lambda_{21} - \Lambda_1 \Gamma_{21})} + \frac{B_{11}^\prime \lambda_1 \lambda_{31} E(1,3;1)}{\nu_1 \nu_2 (\Lambda_{31} - \Lambda_1 \Gamma_{31})(\Gamma_{21} \Lambda_{31} - \Gamma_{31} \Lambda_{21})} \]

\[ + \frac{B_{11}^\prime \lambda_1 \lambda_{31} E(1,2;1)}{\nu_1 \nu_2 (\Lambda_{31} - \Lambda_1 \Gamma_{31})(\Gamma_{31} \Lambda_{21} - \Gamma_{21} \Lambda_{31})} \quad (A.11) \]
where,
\[
\lambda_{kj} = \frac{\lambda_k}{v_k} - \frac{\lambda_j}{v_j}, \quad \Gamma_{kj} = \frac{1}{v_k} - \frac{1}{v_j}, \quad \beta_{ij} = \begin{cases} \\ \Lambda_i, & i=j \\
 \frac{\Lambda_i j}{\Gamma_{ij}}, & i=j \\
 \end{cases}
\]

\[
\gamma_{ijk} = 1 + 4\kappa(\lambda_k - \beta_{ij})/v_k, \quad \Lambda_i = k_i + \lambda_i
\]

Here, (A.9), (A.10) and (A.11) are almost identical to (5.29), (5.30) and (5.31) formally. The only differences are seen in the definition of some parameters, \(\beta_{ij}, \beta'_{ij}, \Lambda_i\). As for the exponential release, (A.9) to (A.11) are also the solution for that if all \(k_i\) is set as \(k\) for \(i = 1, 2, 3\).
A.1.5 Program listing of MGRATO3 (UCB-NE-10)
**PROGRAM MGRATC3(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

**FUNCTION MGRAT03(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

************
* *
* MGRAT03 *
* *
************

A COMPUTER CODE FOR EVALUATING
MIGRATION OF RADIONUCLIDES IN GEOSPHERE

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DATE: MARCH 25, 1980

COMMON/CAL1/BC(3,3),GC(3,3),RC(3,3),C(12),FN(3),FX(3),TN(3)
COMMON/CAL2/ GS(3,3),BT(3,3),R(3),V(3),FK,XG,ZZ,LBC,IER
COMMON/CAL3/ ANAME(3),AT(3),CCEFK(3),CURI(3),HLIFE(3),PR(3)
+RCGW(3),DIF,VEL,TL,TS1,TL1

COMMON/CAL4/ CAA(3),CNB(3),CVC(3)

DIMENSION RR(3),CN(3),CX(3)

DATA QM9/7H
CC 1 I=1,3
DC 1 J=1,3
BC(I,J)= C.
BT(I,J)= 0.
GC(I,J)= C.
RC(I,J)= C.
CC 1 K=1,3
GS(I,J,K)= C.

1 CONTINUE
5 CONTINUE
READ(5,501) ICRL,IA,IB,IC,IO,IE,IF
IF(ICRL.EQ.99) GO TO 480
IF(IA.EQ.0) GC TC 10
INLC= IA
READ(5,502) (ANAME(1),HLIFE(1),CURI(1),CCEFK(1),RCGW(1),PR(1),
+ I=1,IA)
10 IF(IB.EQ.C) GO TO 20
READ(5,501) ICIN,ICOUT,ICAL,IDIV,IBC
20 IF(IC.EQ.0) GC TC 30
READ(5,503) DIF,VEL,FLW
30 IF(IE.EQ.0) GO TO 40
READ(5,504) IREL,TS,TL
40 IF(IE.EQ.0) GO TO 50
READ(5,505) IT,ICT,TL
**PROGRAM MGRATC3(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

50 IF(F.EQ.0) GC TC 60
READ(5,505) IZ,IDZ,Z1
60 CONTINUE
IF(IREL.EQ.3) GC TC 80
DO 70 I=1,INLC
70 PR(I)= TL
EC CONTINUE

**ATOMIC CONSTANTS**

AK= 1.16E13E18
FK= DIF/VEL
VMIN= 1.ELCC
VMAX= C.
DC 100 K=1,INUC
A= ALOG(2.*1/HLIFE(K)
B= PR(K)
R(K)= A
RX(K)=A
TN(K)= B
V(K)= VEL/CCEF(K)
AT(K)= CURIN(K)
IF(IDIN.EQ.1) AT(K)= AT(K)/A*AK
IF(VMIN.GT.V(K)) VMIN= V(K)
IF(VMAX.GT.V(K)) VMAX= V(K)
FR(K)= A
IF(IREL.NE.1) PR(K)= A+1./B

100 CONTINUE
BC(1,1)= AT(1)
E1= EXP(-R(1)*TS)
WA= BC(1,1)*E1
CN(1)= WA
IF(IDIN.EQ.1) CN(1)= CN(1)*R(1)/AK
IF(INUC.NE.1) GC TO 102
ANAME(2)= CNA
ANAME(3)= QA
GC TC 110

102 CONTINUE
R12= R(1)-R(2)
XA= -R(1)/R12
BC(2,1)= AT(1)*R(1)/R12
BC(2,2)= AT(2)-BC(2,1)
E2= EXP(-F(Z)*TS)
WA= BC(2,1)*E1+BC(2,2)*E2
CN(2)= WA
IF(IDIN.EQ.1) CN(2)= CN(2)*R(2)/AK
IF(INUC.NE.2) GC TC 104
ANAME(3)= CNA
GC TC 110

104 CONTINUE
R23= R(2)-R(3)
R31= R(3)-R(1)
RRR= AT(1)*R(1)*R(2)
XB= -R(2)/R23
XC= -R(1)*R(2)/(R12*R31)
XD= -R(1)*R(2)/(R12*R23)
XE= -R(1)*R(2)/(R31*R23)
E3= EXP(-TS*R(3))
**FRCGRAM**

**MGFATOC3** (INPUT, OUTPUT, TAPE5=INPUT, TAPE6=OUTPUT)**

BC(3,1) = -RRR/(R12*R31)
EC(3,2) = -RRR/(R12*R23)-AT(2)*R(21/R23
BC(3,3) = -RRR/(R23*R31)+AT(2)*R(2)/R23+AT(3)

CWA = BC(3,1)*E1+BC(3,2)*E2+BC(3,3)*E3
CN(3) = CWA

IF(IDINC.EQ.1) CN(3) = CN(3)*R(3)/AK

110 CONTINUE

CC 112 K=1, NUC
AT(K) = CN(K)/(FR(K)*FLH)

112 IF(IDINC.EQ.1) AT(K) = AT(K)*AK/R(K)

C

PRINT CLT GF INITIAL CONDITIONS

C

CALL TITLE1 (INUC, IFEL, ICAL, IDINC, CN)

BC(1,1) = AT(1)
BT(1,1) = RR(1)
GS(1,1,1) = GMF(1,1,1)
IF(INUC.EQ.1) GC TC 120
R12 = RR(1)-RR(2)
BC(2,1) = -AT(1)*R(1)/R12*PR(1)/PR(2)
BC(2,2) = AT(2)-BC(2,1)
FC(1,2) = F(1)/V(1)-F(2)/V(2)
GC(1,2) = 1.0/V(1)-1.0/V(2)
BT(1,2) = RC(1,2)/GC(1,2)
BT(2,2) = RR(2)
GS(1,2,1) = GMF(1,2,1)
GS(1,1,2) = GMF(1,1,2)
GS(2,2,2) = GMF(2,2,2)
GS(1,2,2) = GMF(1,2,2)
RV1 = R11/V(1)
C(1) = RV1/(RC(1,2)+FR(1)*GC(1,2))
IF(INUC.EQ.2) GC TC 120
R23 = RR(2)-RR(3)
R31 = RR(3)-RR(1)
RRR = AT(1)*R(1)*R(2)*PR(1)/PR(3)
EC(3,1) = -RRR/(F12*R31)
BC(3,2) = -RRR/(R12*R23)-AT(2)*R(2)/R23*PR(2)/PR(3)
BC(3,3) = -RRR/(R23*R31)+AT(2)*R(2)/R23*PR(2)/PR(3)+AT(3)
FC(2,1) = -RC(1,2)
RC(3,1) = R(3)/V(3)-R(1)/V(1)
RC(1,3) = -RC(3,1)
RC(3,2) = F(3)/V(3)-F(2)/V(2)
RC(2,3) = -RC(3,2)
GC(2,1) = -GC(1,2)
GC(3,1) = 1.0/V(3)-1.0/V(1)
GC(1,3) = -GC(3,1)
CC(3,2) = 1.0/V(3)-1.0/V(2)
GC(2,3) = -GC(3,2)
BT(1,3) = RC(1,3)/GC(1,3)
BT(2,3) = RC(2,3)/GC(2,3)
BT(3,3) = RR(3)
GS(1,3,1) = GMF(1,3,1)
GS(2,3,2) = GMF(2,3,2)
GS(1,3,1) = GMF(1,3,1)
GS(1,3,2) = GMF(1,3,2)
GS(2,2,3) = GMF(2,2,3)
GS(2,3,1) = GMF(2,3,1)
GS(3,3,3) = GMF(3,3,3)
**PROGRAM MGRATC3(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

RV2 = F(21)/V(2)
RV3 = RV4*RV2
C(2) = RV2/(RC(2,3)-RR(2)*GC(2,3))
C(3) = RV2/(RC(2,3)-FR(1)*GC(2,3))
C(4) = RV1/(RC(1,3)-FR(1)*GC(1,3))*C(3)
CA = RV3/((GC(1,3)*RC(2,3)-GC(2,3)*RC(1,3))
CB = RV3/((GC(3,2)*RC(1,2)-GC(1,2)*RC(3,2))
CC = RV3/((GC(3,1)*RC(2,1)-GC(2,1)*RC(3,1))
CA = GC(1,3);CA/((RC(1,3)-RR(1)*GC(1,3))
C(6) = GC(2,3);CA/((RC(2,3)-RR(1)*GC(2,3))
C(7) = RV2/(RC(3,2)-FR(1)*GC(3,2))*C(1)
C(8) = GC(3,2)*CB/((RC(3,2)-RR(1)*GC(3,2))
C(9) = GC(1,2)*CB/((RC(1,2)-RR(1)*GC(1,2))
C(10) = RV3/(RC(3,1)-RR(1)*GC(3,1))/(RC(2,1)-RR(1)*GC(2,1))
C(11) = GC(3,1);CC/((RC(3,1)-RR(1)*GC(3,1))
C(12) = GC(2,1)*CC/((RC(2,1)-RR(1)*GC(2,1))

120 CONTINUE
IF(ICAL.EQ.1) CC TO 440

C FOR FIXED TIME

C
FD = S./FLCAT(TZ)
DO 420 M=1,17
TZ = T1*10.**(M-1)
KM = 1
GO TO 402
401 CONTINUE
TZ = T2*5.C
KM = 0
402 CONTINUE
CALL TITLE2(ICAL,IDCUT,TZ)
DO 416 N=1,17
LC = 416 L=1,17
FL = L
Z2 = Z1*10.**(N-1)*1.FD*(FL-1)
IER = 0
CALL CALC(INUC,C,T2,Z2)
IF(IER.EQ.1) GC TC 417
DC 403 LL=1,INUC
403 CN(LLL) = FN(LLL)
IF(IREL.EQ.1) GC TC 407
IF(T2.LE.TL) GC TC 407

C BAND RELEASE

C
IER = C
CALL CALC(INUC,1,TZ,Z2)
IF(IER.EQ.1) GC TC 417
DC 405 LL=1,INUC
405 CN(LLL) = CN(LLL)-FN(LLL)
407 CONTINUE
CMAX = C
CO 405 LL=1,INUC
WA = CN(LLL)/AT(1)
IF(CMAX.LT.WA) CMAX = WA
CNA(LLL) = WA
409 CN(LLL) = WA*R(LLL)/F(1)
IF(IDOUT.NE.1) GC TC 413
CO 411 LL=1,INUC
**PRGGRAM MGRATC3(INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)**

CNA(LL) = R(LL)*C(nLL)*FLW/AK
411 CNE(LL) = CNA(LL)/FCGW(LL)
413 CONTINUE
ZMAX = V*IAN*T2
IF(ZMAX.GT.Z2) GC TC 415
IF(CMAX.LT.1.E-10) GO TO 417
415 CONTINUE
CALL PRINTR(INLC,ICLT,ICAL,Z2)
416 CONTINUE
417 CONTINUE
IF(IDIV.EQ.1) GC TC 420
IF(KM.EQ.1) GO TO 401
420 CONTINUE
GC TC 5
C
C FCR FIXED LCCATION
C
440 CONTINUE
FD= 9./FLCAT(IDT)
CO 460 M=1,IZ
Z2 = Z1*10.**((M-1)
KM= 1
GO TO 442
441 CONTINUE
Z2 = Z2*5.C
KM= 0
442 CONTINUE
CALL TITLE2(ICAL,IDCUT,Z2)
TX= 0.
CA= 0.
QB1 = 0.
CB2= 0.
QC1= 0.
QC2= 0.
QC3= 0.
DC 454 =1,IT
CO 454 =1,IDT
FL= L
T2= TL*1C.**((N-1)*(1.+FD*(FL-1.))
C
IER= 0
CALL CALC(INUC,C,T2,Z2)
IF(IER.EQ.1) GC TC 456
DC 443 LL=1,INUC
443 CNLL) = FN(LLL)
IF(IDREL.NE.1) GC TC 446
IF(T2.LE.TL) GC TC 446
C
C BAND RELEASE
C
IER= 0
CALL CALC(INUC,1,T2,Z2)
IF(IER.EQ.1) GO TO 456
EC 444 LL=1,INUC
444 CNLL) = CALLI-FN(LLL)
446 CONTINUE
DET= T2-TX
IF(IDCUT.EQ.C) GC TC 1000
**FRCGRAM MGRAT03 (INPUT, OUTPUT, TAPE5=INPUT, TAPE6=OUTPUT)**

E1 = EXP(-R(1)*DET)

CA = CA*E1*(1.0-E1)*CN(1)/R(1)

CX(1) = QA

IF(INUC.EQ.1) G0 TO 1000

E2 = EXP(-R(2)*DET)

QB1 = QB1*E1*(1.0-E1)*CN(1)*XA/R(1)

QB2 = QB2*E2*(1.0-E2)*CN(2)*CN(1)*XE/R(2)

CX(2) = QB1+QB2

IF(INUC.EQ.2) G0 TO 1000

E3 = EXP(-R(3)*DET)

QC1 = QC1*E1*(1.0-E1)*CN(1)*XC/R(1)

QC2 = QC2*E2*(1.0-E2)*CN(2)*CN(1)*XD/CN(1)/R(2)

QC3 = QC3*E3*(1.0-E3)*CN(3)*XB/CN(2)*XE/CN(1)/R(3)

CX(3) = QC1+QC2+QC3

1000 CONTINUE

CMAX = 0.0

DG 447 LL=1, INLC

WA = CN(LL)/AT(1)

IF(CMAX.LT.WA) CMAX = WA

CNA(LLL) = WA

447 CNB(LLL) = WA*R(LLL)/R(1)

IF(IDCLT.LE.1) GC TC 450

DO 448 LL=1, INLC

CNA(LLL) = R(LLL)*CN(LLL)*FLW/AK

WA = R(LLL)*CX(LLL)*FLW/AK

CNC(LLL) = WA/RCGW(LLL)

448 CNE(LLL) = CNA(LLL)/FRCGW(LLL)

450 CONTINUE

TX = T2

TMAX = Z2/VMIN+TL

IF(TMAX GT T2) GC TC 452

IF(CMAX LT 1.0E-10) GC TO 456

452 CONTINUE

TS2 = T2+TS

CALL PFINTR(INUC, ICOUT, ICAL, TS2)

454 CONTINUE

456 CONTINUE

IF(IDIV.EQ.1) GC TO 460

IF(KM.EQ.1) GC TC 441

460 CONTINUE

GC TC 5

480 STOP

5C1 FORMAT(712)

5C2 FCFMAT(A7,5F8.0)

5C3 FORMAT(3F8.0)

504 FCFMAT(I1,2F8.0)

5C5 FCFMAT(I1,3,2F8.0)

END
**SUBROUTINE CALC(INUC,IK,T,Z)**

SUBROUTINE CALC(INUC,IK,T,Z)
COMMON/CA1/BC(3,3),GC(3,3),RC(3,3),C(12),FN(3),R(3),TL(3)
DIEMNSION TM(3),B(3,3)
DC IC I=1,INLC
10 TM(I)= 1.0
  IF(IK.EQ.0) GC TO 30
  DO 20 I=1,INLC
20 TM(I)= EXP(-R(I)*TL(I))
30 CCNTINUE
  DO 40 I=1,INLC
  DO 40 J=1,INUC
40 B(I,J)= BC(I,J)*TM(J)
  DO 50 I=1,INLC
50 TM(I)= T
  IF(IK.EQ.0) GC TC 70
  DO 60 I=1,INLC
60 TM(I)= T-TL(I)
70 CCNTINUE
   TA = TM(1)
    XA = EFN(1,1,1,T,TA,Z)
    FN(1) = E(1,1)*XA
    IF(INUC.EQ.1) GO TO 100
   TB = TM(2)
    XB = EFN(2,2,2,TA,Z)
    XC = EFN(1,1,2,TA,Z)
    XD = EFN(1,2,2,TA,Z)
    XF = EFN(1,2,1,TA,Z)
    FN(2) = B(2,2)*XB+B(2,1)*XC+B(1,1)*C(1)*(XC-XA-XD+XE)
    IF(INUC.EQ.2) GC TO 100
   TC = TM(3)
    XF = EFN(3,3,3,TC,Z)
    XC = EFN(2,2,3,TC,Z)
    XH = EFN(1,3,3,TC,Z)
    XI = EFN(2,3,3,TC,Z)
    XJ = EFN(2,3,2,TC,Z)
    XK = EFN(1,3,3,TC,Z)
    XL = EFN(1,3,1,TC,Z)
    FN(3) = B(3,3)*XF+XG+B(3,1)*XH
        + B(2,2)*C(2)*(XC-XI-XB+XJ)+B(2,1)*C(3)*(XH-XI-XC+XJ)
        + B(1,1)*(XH*C(4)-XK*C(5)+XI*C(6)+XC*C(7)-XJ*C(8)+XD*C(9)
        + XA*C(10)-XL*C(11)+XE*C(12))
100 RETURN
END
**SUBROUTINE GALLS(FNM,A,B,Q)**

SUBROUTINE GALLS(FNM,A,B,Q)
G = C.
AB = (A+B)/2.
BA = E-A

10 FCINT GAUSSIAN-LEGENDRE QUADRATURE FORMULA

C = .4665573*BA
Q = .033333367*(FNM(AB+C)+FNM(AB-C))
C = .4321517*BA
Q = Q+.07472567*(FNM(AB+C)+FNM(AB-C))
C = .3399712*BA
Q = Q+.1556526*(FNM(AB+C)+FNM(AB-C))
C = .2166677*BA
Q = Q+.1040628*(FNM(AB+C)+FNM(AB-C))
C = .0744317*BA
Q = Q+.0477601*(FNM(AB+C)+FNM(AB-C))
Q = C*BA
RETURN
ENC

**SUBROUTINE PRINR(INUC,IDOUT,ICAL,TZ)**

SUBROUTINE PRINR(INUC,IDOUT,ICAL,TZ)
COMMON/ICAL,10,ICAL,TZ
COMMON/ICAL,CNA(3),CNB(3),CNC(3)
DIMENSION LISA1(3),LISA2(3),LISA3(3),LYAN1(3),LYAN2(3),LYAN3(3)
DATA LISA1/15X,1PE10.3,2(4X,1E10.3,20X)/
+ LISA2/3CH(15X,1PE10.3,2(4X,2E10.3,10X))/
+ LISA3/2CH(15X,1PE10.3,2(4X,3E10.3))/
+ LYAN1/26H(15X,1PE10.3,3(4X,1E10.3,20X))/
+ LYAN2/3CH(15X,1PE10.3,3(4X,2E10.3,10X))/
+ LYAN3/26H(15X,1PE10.3,3(4X,3E10.3))/
IF(ICAL.NE.1) GC TC 10
IF(IDOUT.EQ.1) GO TO 100
10 CCNTABLE
IF(INUC.EQ.2) GC TC 20
IF(INUC.EQ.3) GO TO 30
WRITE(6,LISA1) TZ,CNA(I),CNB(I)
RETURN
20 WRITE(6,LISA2) TZ,(CNA(I),I=1,2),(CNB(I),I=1,2)
RETURN
30 WRITE(6,LISA3) TZ,(CNA(I),I=1,3),(CNB(I),I=1,3)
RETURN
1CC CCNTABLE
IF(INUC.EQ.2) GO TO 120
IF(INUC.EQ.3) CC TO 130
WRITE(6,LYAN1) TZ,CNA(I),CNB(I),CNC(I)
RETURN
120 WRITE(6,LYAN2) TZ,(CNA(I),I=1,2),(CNB(I),I=1,2),(CNC(I),I=1,2)
RETURN
130 WRITE(6,LYAN3) TZ,(CNA(I),I=1,3),(CNB(I),I=1,3),(CNC(I),I=1,3)
RETURN
END
**SUBROUTINE TITLE1(INUC, IREL, ICAL, ICIN, CAN)**

SUBROUTINE TITLE1(INUC, IREL, ICAL, ICIN, CNX)
COMMON/CALZ2/ GS(3,3), BT(3,3), R(3,3), V(3), FK, XG, ZZ, IEC, IER
COMMON/CAL3/ ANAME(3), AT(3), CCEFK(3), CURIN(3), HLIFE(3), PR(3)
   + , RCGW(3), CIF, VEL, FLW, TS, T1, Z1, IT, IZ
DIMENSION A(3), B(2), D(2), CNX(3), F(2)
DATA A(1), A(2), A(3), B(1), B(2), D(1), D(2) / 7HBAND RL, 7HXP FL, 7HPREF 
   + RL, 7HFIXED Z, 7HFIXED T, 7HCL/M3, 7H (N/M3)/( 
DATA F(1), F(2) / 7HSLCE, 7HCONC B /
WRITE(6, 600) INUC

600 FCMAT(1H1, 40X, * MIGRATION CF *, T, 1, MEMBER CHAIN THRU A 
   + SORBING MEDIUM ---, *, 55X, --- INPUT DATA ---*)
WRITE(6, 601) VEL, CIF, FLW

601 FCMAT(40X, * FCERC DATA, *, 55X, VELOCITY DEPNSN FLOWRATE*, /
   + 57X, *(M/Y) (M2/Y) (M3/Y1), *, 55X, 1PEB.2, 2(2X, 1E8.2))
WRITE(6, 602) A(IREL, TS

602 FCMAT(40X, * FCER DATA, *, 55X, RELEASE MCDE *, 55X, LEA 
   + C TIME *, 1PEB.2, *(Y)*)
C = F(1)
IF (ICAL .NE. 1) C = B(2)
G = F(1)
IF (ICAL .EQ. 1) G = F(2)
WRITE(6, 603) C, G, T1, IT, Z1, IZ

   + 55X, T FROM *, 1PEB.2, *(Y) TC X10---, 1I, 55X, Z FROM *, 
   + 8E.2, *(M) TC X10---, 1I)
WRITE(6, 604) (ANAME(I), HLIFE(I), R(I), COEFP(I), V(I), I=1, INUC)

   + DECAY CST*, 6X, Ki VALUE*, 7X, VELOCCITY*, 22X, *(-)*, 1I, X, *(Y) *, 10X, 
   + *(1/Y) *, 10X, *(1), 12X, *(M/Y) +, 3(2X, A7, 1P4E15.3, 1/))
E = D(1)
IF (ICAL .NE. 1) E = D(2)
WRITE(6, 605) E, E, (ANAME(I), CURIN(I), CNX(I), PR(I), RCGW(I), I=1, INUC)

   + 6X, RCG WATER*, 22X, *(-)*, 1I, X, A7, 8X, A7, 8X, *(Y) *, 1I, X, *(1/M3) , 1/ 
   + (20X, A7, 1F4E15.3, 1/)
WRITE(6, 606)

606 FCMAT(1H1)
RETURN
END

**SUBROUTINE TITLE2(ICAL, ICUT, T)**

SUBROUTINE TITLE2(ICAL, ICUT, T)
COMMON/CAL3/ ANAME(3), AT(3), CCEFK(3), CURIN(3), HLIFE(3), PR(3)
   + , RCGW(3), DIF, VEL, FLW, TS, T1, Z1, IT, IZ
DATA A, B/ 5H T, 5T Z = /
C = A
IF (ICAL .EQ. 1) C = B
IF (ICUT .EQ. 1) GO TO 1C
WRITE(6, 650) C, T, (ANAME(I), I = 1, 3), (ANAME(I), I = 1, 3)
RETURN
10 WRITE(6, 650) C, T, (ANAME(I), I = 1, 3), (ANAME(I), I = 1, 3)
   , (ANAME(I), I = 1, 3)
RETURN
650 FCMAT(40X, 1PE10.3, 14X, 3(2X, A7, 1X), 4X, 3(2X, A7, 1X)
   + 4X, 3(2X, A7, 1X))
END
**FUNCTION EFN(I,J,K,T,Z)**

FUNCTION EFN(I,J,K,T,Z)
CALL GS(3,3,3),BT(3,3),R(3),V(3),FK,XG,ZZ,IBC,IER
EXTERNAL FAM
ZZ= Z
XT= V(K)*T
XL= GS(I,J,K)
IF(XL.LE.0.0) GO TO 100
IF(FK.LT.1.0) GC TC 200
IF(Z.LT.XT) GC TC 200
GO TO 200
100 CONTINUE
C C NUMERICAL INTEGRATION
C AL= 0.0
AP= SQRT(XT/FK/4.0)
IF(IBC.EQ.1) AP= SQRT(4.*FK*XT)/ZZ
N= 20
RES= 0.0
AD= (AP-AL)/FLCAT(N)
DO 110 LL= 1,N
AL= AC*FLCAT(LL-1)
AH= AD*FLCAT(LL)
110 CONTINUE
RES= RES+RESLLT
IF(IER.EQ.1) GC TC 300
XA= -BT(I,J)*T
EFN= EXP(XA)*RES*2./1.7724538
RETURN
200 CONTINUE
C C RATIONAL APPROXIMATION
C AZ= ABS(Z)
XG=SQRT(XL)
XB= SQRT(4.*FK*XT)
XC= BT(I,J)
FK2= 2.*FK
X= Z/FK2-XC*T-AZ*XG/FK2
IF(X.GT.650.0) GC TC 300
X= EXP(X)
Y= (AZ-XT*XG)/XE
Y= 1.0-ERF(Y)
X= X*Y
Y= (AZ+XT*XG)/XB
W= Z/FK2-XC*T+AZ*XG/FK2-Y*Y
IF(W.GT.650.0) GO TC 300
W= EXP(W)*(1.-ERF(YI))
EFN= (X-H)/XG/2.
IF(IBC.EQ.1) EFN= (X+H)/2.
RETURN
C C ERROR
C 300 EFN= C.
RETURN
END
**FUNCTION ERF(XX)**

FUNCTION ERF(XX)

C
C ERF = 2/SQRT(PI)*INTEGRAL CF EXP(-T*T) FRCP . O TC X.
C USING AN APPROXIMATION DUE TO HASTINGS. ABSOLUTE ERROR ABOUT 3E-7
C
DIMENSION A(6)
DATA A/* .000430638, .002785672, .001520143, .0052705272, .0422820123
+ .075230784 */
XX = ABS(XX)
T = A(1)*X
DC 10 I=2,6
T = (T+A(I))*X
10 CONTINUE
T = 1./(T+1.)
ERF = 1.-T**16
IF(XX.LT.0.) ERF = -ERF
RETURN
END

**FUNCTION FNM(X)**

FUNCTION FNM(X)
C$**CAL2/ GS(3,3,3), BT(3,3), R(3), V(3), FK, XG, ZZ, IBC, IER
IF(IBC.EQ.1) GC TC 10
XA = ZZ/(4.*FK*X)
XB = XG*X*X+XA*X
XB = ZZ/FK/2.*XB
IF(XB.GT.650.) GO TO 20
FNM = EXP(XB)
RETURN
10 XA = ZZ*X/(4.*FK)
XB = 1./(X*X)+XA*X
XB = ZZ/FK/2.-XB
IF(XB.GT.650.) GC TO 20
FNM = 1./(X*X)*EXP(XB)
RETURN
C
C ERROR
C
20 FNM = 0.
IER = 1
RETURN
ENC

**FUNCTION GMF(I, J, K)**

FUNCTION GMF(I, J, K)
COMMON/CAL2/ GS(3,3,3), BT(3,3), R(3), V(3), FK, XG, ZZ, IBC, IER
GMF = 1.0+4.0*FK*(R(K)-BT(I,J))/V(K)
RETURN
ENC
A.1.6 Input data card instruction
### INPUT DATA CARD INSTRUCTION

1st CARD

<table>
<thead>
<tr>
<th>I2</th>
<th>2</th>
<th>99; End of Input Data</th>
<th>Otherwise: Case Study</th>
</tr>
</thead>
<tbody>
<tr>
<td>I2</td>
<td>2</td>
<td>Skip Nuclide Data</td>
<td>1 or 2 or 3: Number of Nuclide in a Chain</td>
</tr>
<tr>
<td>I2</td>
<td>3</td>
<td>Skip 5th Card</td>
<td>Otherwise: Read 5th Card</td>
</tr>
<tr>
<td>I2</td>
<td>6</td>
<td>Skip 6th Card</td>
<td>Otherwise: Read 6th Card</td>
</tr>
<tr>
<td>I2</td>
<td>7</td>
<td>Skip 7th Card</td>
<td>Otherwise: Read 7th Card</td>
</tr>
<tr>
<td>I2</td>
<td>8</td>
<td>Skip 8th Card</td>
<td>Otherwise: Read 8th Card</td>
</tr>
<tr>
<td>I2</td>
<td>9</td>
<td>Skip 9th Card</td>
<td>Otherwise: Read 9th Card</td>
</tr>
</tbody>
</table>

2nd Card

<table>
<thead>
<tr>
<th>I2</th>
<th>4</th>
<th>Name of Nuclide</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>5</th>
<th>Half-Life of Nuclide (yr)</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>6</th>
<th>Initial Amount of Nuclide (g) or (Atom)</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>7</th>
<th>Retardation Factor of Nuclide (-)</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>8</th>
<th>Radioactivity Conc. Guide (g/m³ - H₂O)</th>
</tr>
</thead>
</table>

| I2 | 9 | Time Constant for Pref. Release (yr) |

5th Card

<table>
<thead>
<tr>
<th>I2</th>
<th>1</th>
<th>Dimension of INPUT: C₄</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>2</th>
<th>Dimension of OUTPUT: C₄</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>3</th>
<th>Mode of Calculation: Fixed t</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>4</th>
<th>Interval of Fixed t or t: Every 5</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>I2</th>
<th>5</th>
<th>Transport Eq. used: 1: Conc. BC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Card</td>
<td>Description</td>
<td></td>
</tr>
<tr>
<td>------</td>
<td>-----------------------------------------------------------------------------</td>
<td></td>
</tr>
<tr>
<td>6th</td>
<td>F6.0  Dispersion Coefficient (m²/yr)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F8.0  Velocity of Water (m/yr)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F8.0  Flow Rate (m³/yr)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F8.0  Time of Initial Release after Emplacement (yr)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F8.0  Leach Time or Time Constant for Exponential Release (yr)</td>
<td></td>
</tr>
<tr>
<td>8th</td>
<td>Steps of Time</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Interval of Time, Select 45, 90, ...., 900</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F8.0  Initial Time for Calculation (yr)</td>
<td></td>
</tr>
<tr>
<td>9th</td>
<td>Steps of Distance</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Interval of Distance, Select 45, 90 , ...., 900</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F8.0  Initial Distance for Calculation (m)</td>
<td></td>
</tr>
</tbody>
</table>
A.1.7 Sample of OUTPUT
--- MIGRATION OF 3-MEMBER CHAIN THRU A SORBING MEDIUM ---

--- INPUT DATA ---

1. HYDRO DATA

VELOCITY DISPERSION

\[
\begin{array}{c}
\text{(M/Y)} \\
1.00E+01
\end{array}
\quad
\begin{array}{c}
\text{(M2/Y)} \\
1.00E+01
\end{array}
\quad
\begin{array}{c}
\text{(M3/Y)} \\
1.00E+01
\end{array}
\]

2. RELEASE DATA

RELEASE MODE  PREF RL

LEAD TIME  1.00E+03 (Y)

3. CALCULATION DATA

FIXED Z

SOURCE CONDITION

T FROM 1.00E+03 (Y) TO x10--4

Z FROM 1.00E+01 (M) TO x10--1

4. NUCLIDES DATA

\[
\begin{array}{|c|c|c|c|c|}
\hline
\text{NUCLIDE} & \text{HALF LIFE} & \text{DECAY CST} & \text{KI VALUE} & \text{VELOCITY} \\
& \text{(Y)} & \text{(1/Y)} & \text{(-)} & \text{(M/Y)} \\
\hline
\text{U 234} & 2.440E+05 & 2.841E-06 & 1.430E+04 & 6.993E-04 \\
\text{TH 230} & 7.700E+04 & 9.002E-06 & 5.000E+04 & 2.000E-04 \\
\text{RA 226} & 1.600E+03 & 4.332E-04 & 5.000E+02 & 2.000E-02 \\
\hline
\end{array}
\]

\[
\begin{array}{|c|c|c|c|c|}
\hline
\text{NUCLIDE} & \text{CCNC. IA} & \text{CCNC. ST} & \text{LEACH TIME} & \text{RCG WATER} \\
& \text{(Cl/M3)} & \text{(Cl/M3)} & \text{(Y)} & \text{(Cl/M3)} \\
\hline
\text{U 234} & 1.146E+00 & 1.143E+00 & 1.000E+04 & 3.000E-05 \\
\text{TH 230} & \text{C.} & 1.026E-02 & 1.000E+04 & 2.000E-06 \\
\text{RA 226} & \text{C.} & 1.936E-03 & 1.000E+04 & 3.000E-08 \\
\hline
\end{array}
\]
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