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EFFECT OF MINERALIZATION REACTION  
ON THE RADIONUCLIDE TRANSPORT  
THROUGH GEOLOGIC MEDIA

November 1980

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Effect of Mineralization Reaction on the Radionuclide  
Transport Through Geologic Media

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When the radioactive nuclear-power waste which was emplaced in a geologic repository begins to release from repository, dissolve into ground water and migrate in geologic media, the irreversible sorption process would arise from the mineralization reaction of the sorbed nuclides. In the present report we have proposed the migration model with irreversible mineralization. In addition the recursive solution of the transport equation with equilibrium sorption process and irreversible mineralization of a portion of the sorbed radionuclides has been developed. The explicit solution has been developed and demonstrated by numerical examples for three member decay chain and for arbitrary nuclide source and step release.

Keywords; Mineralization, Radionuclide, Geologic Media,  
Waste, Repository, Irreversible Sorption, Migration,  
Recursive Solution, Explicit Solution, Ground Water,  
Calculation Models

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地層中の放射性核種の移行におよぼす鉱物化の効果

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地層処分された高レベル放射性廃棄物中の放射性核種が、処分場から浸出し、地下水により地中を移行する際、核種と岩石あるいは核種と地下水中の成分が反応し、反応生成物をつくるいわゆる鉱物化の起こることが考えられる。この研究では、鉱物化を考慮した核種の地下水中から、生物環境に至る核種の移行について、理論的、解析的に予測する計算コードの開発を試みた。具体的には、局所的に平衡吸着過程が成り立つとの仮定のもとに、一部不可逆鉱物化を考慮した核種の地下水中の移行方程式について、リカーシブな解を求め、ついで、任意の核種源の存在とステップリリースの条件の下に、3核種崩壊チェーンのそれぞれの核種についての解析解を求め、典型的な計算例を示した。

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## 1. Introduction

Current risk assessment<sup>(1), (2)</sup>, in which irreversible reactions such as irreversible sorption reaction and mineralization are ignored, may be overestimating their hazard to the public. While the radionuclides migrate in the ground water, they react with elements in ground water and dissolve in aqueous phase with the various chemical forms. These reactants have different equilibrium sorption properties such as parameters of  $k_{di}$ .

On the other hand, at the surface of solid phase, nuclide might be trapped with some mechanism and react with rock.

This is the mineralization reaction.

The purpose of this note is to estimate the effect of mineralization on migration of radionuclide through the geologic media.

## 2. Mass Transport

In this note, we assume that the sorption and chemical reaction process are attained to equilibrium state locally. The sorption equilibrium coefficient for nuclide is assumed as

$$k_{di} = \frac{\text{Concentration of nuclide in solid phase}}{\frac{\text{mass of medium}}{\text{Concentration of nuclide in aqueous phase}}} = \frac{\text{Concentration of nuclide in solid phase}}{\frac{\text{volume of solution}}{\text{Concentration of nuclide in aqueous phase}}}$$

The chemical reaction equilibrium relation is assumed to be given by a linear relation between different chemical species of the nuclide element. The other assumptions made are as follows:

- (1) One dimension
- (2) Dispersion or diffusion-like migration with convection
- (3) Characteristic properties of geologic media are invariant spatially and in time
- (4) Convection and diffusion in solid phase are neglected
- (5) Source term is in the transport equation

(6) Step release mode

(7) Mineralization is a first order rate process

The situation of mineralization is shown in Fig. 1 schematically. The horizontal arrows between squares represent the radioactive decay of the nuclide.

The vertical arrows show chemical reaction and mineralization reaction. The diagonal arrows represent reversible mineralization reaction.

### 2.1 Transport Equation

In this model transport equation for  $i$ -th nuclide can be written as follows

$$\varepsilon \frac{\partial N_i}{\partial t} + \varepsilon V \frac{\partial N_i}{\partial z} - \varepsilon D \frac{\partial^2 N_i}{\partial z^2} = -R_{ad,i} - \lambda_i N_i + \lambda_{i-1} N_{i-1} + f_i(z,t)$$

[in aqueous phase]

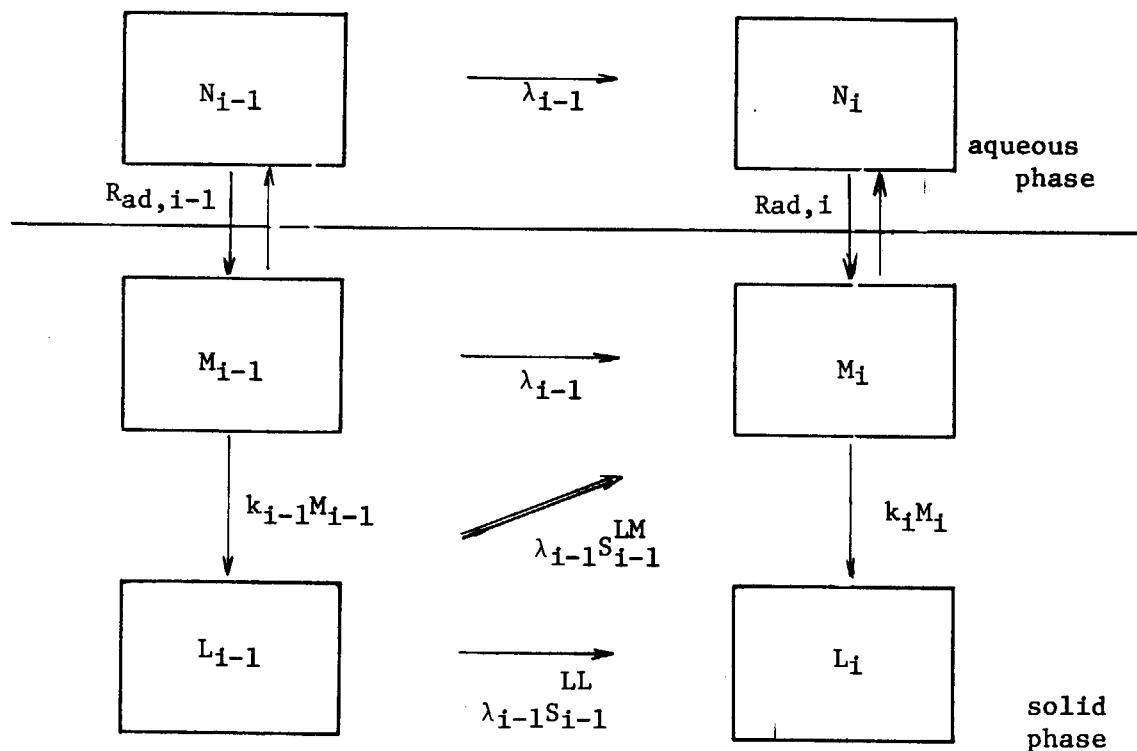
$$(1-\varepsilon) \frac{\partial M_i}{\partial t} = R_{ad,i} - (1-\varepsilon) \lambda_i M_i + (1-\varepsilon) \lambda_{i-1} [M_{i-1} + S_{i-1}^{LM} L_{i-1}] - k_i M_i (1-\varepsilon)$$

$$(1-\varepsilon) \frac{\partial L_i}{\partial t} = -(1-\varepsilon) \lambda_i L_i + (1-\varepsilon) \lambda_{i-1} S_{i-1}^{LL} L_{i-1} + k_i M_i (1-\varepsilon)$$

[in solid phase]

$$R_{ad,i} = k_m (N_i - \frac{1}{k d_i} M_i)$$

$$S_{i-1}^{LM} + S_{i-1}^{LL} = 1$$



$k_{i-1} M_{i-1}$  : reaction rate of mineralization  
 $N_{i-1}$  :  $i-1$  th nuclide in aqueous phase  
 $M_{i-1}$  :  $i-1$  th desorbable nuclide in solid phase  
 $L_{i-1}$  :  $i-1$  th mineralized nuclide in solid phase  
 $\lambda_{i-1}$  : radioactive decay rate coefficient  
 $R_{ad,i-1}$  : chemical reaction rate coefficient  
 $S_{i-1}^{LM}$  : fraction of  $i-1$  th mineralized nuclide L that becomes  $i$  th desorbable nuclide M  
 $S_{i-1}^{LL}$  : fraction of  $i-1$  th mineralized nuclide L that becomes  $i$  th mineralized nuclide L

Fig. 1 Schematic diagram of a decay chain of one species in aqueous phase and two species in solid phase with reversible mineralization

where

$N_i$	nuclide concentration in aqueous phase, $i$ th nuclide (Atom/m <sup>3</sup> )
$M_i$	nuclide concentration in solid phase, $i$ th desorbable nuclide (Atom/m <sup>3</sup> of water)
$L_i$	nuclide concentration in solid phase, $i$ th nuclide impossible to desorb (Atom/m <sup>3</sup> of water)
$\epsilon$	porosity fraction ( - )
$V$	groundwater flow velocity (m/yr)
$D$	dispersion coefficient (m <sup>2</sup> /yr)
$f_i(z,t)$	source term of $i$ th nuclide in aqueous phase (Atom/m <sup>3</sup> yr)
$k_m$	rate coefficient of sorption process ( - )
$k_{di}$	sorption equilibrium coefficient (ml/g)

From equation (1) with the assumption of local equilibrium state, the final governing equation in aqueous phase is

$$\left. \begin{aligned} K_i \frac{\partial N_i}{\partial t} + V \frac{\partial N_i}{\partial z} - D \frac{\partial^2 N_i}{\partial z^2} = - \lambda_i^* K_i N_i + \lambda_{i-1} K_{i-1} N_{i-1} + \frac{1-\epsilon}{\epsilon} S_{i-1}^{LM} L_{i-1} + f_i(z,t) \\ \text{here } \lambda_i^* = \lambda_i + \frac{k_i(1-\epsilon)k_{di}}{\epsilon K_i} \\ K_i = 1 + \frac{1-\epsilon}{\epsilon} k_{di} \end{aligned} \right\} (2)$$

where

$K_i$  Overall sorption coefficient ( - )

## 2.2 Solution of the Governing Equation

The solution of equation (2) can be obtained with the help of Green's function. When  $f_i(z,t)$  is expressed as  $f_i(z,t)/K_i = f_i(t) \cdot \delta(z)$  the recursive solution is given by

$$\left. \begin{aligned} N_i(z,t) = \int_0^t f_i(t-\theta) \frac{e^{-\lambda_i \theta}}{\sqrt{4\pi K_i \theta}} \exp\left[-\frac{(z-v_i \theta)^2}{4K_i \theta}\right] d\theta \\ + \frac{\lambda_{i-1} K_{i-1}}{K_i} \int_0^t \int_{-\infty}^{\infty} N_i(\xi, t-\theta) \frac{e^{-\lambda_i \theta}}{\sqrt{4\pi K_i \theta}} \exp\left[-\frac{z-v_i \theta-\xi}{4K_i \theta}\right] d\xi d\theta \end{aligned} \right\} (3)$$

In the case of decaying step release mode,  $N_i(t)$  is expressed by Bateman equation.

Source term  $f_i(t)$  for three members are;

$$\begin{aligned} f_1(t) &= v_1 N_1(t) \\ &= v_1 N_1^0 e^{-\lambda_1 t} \\ &= v_1 B_{11} e^{-\lambda_1 t} \end{aligned}$$

$$\begin{aligned} f_2(t) &= v_2 N_2(t) \\ &= v_2 \left[ (N_2^0 + N_1^0 \frac{\lambda_1}{\lambda_1 - \lambda_2}) e^{-\lambda_2 t} + N_1^0 \frac{\lambda_1}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} \right] \\ &= v_2 [B_{22} e^{-\lambda_2 t} + B_{21} e^{-\lambda_1 t}] \end{aligned}$$

$$\begin{aligned} f_3(t) &= v_3 N_3(t) \\ &= v_3 \left[ (N_3^0 + \frac{N_2^0 \lambda_2}{\lambda_2 - \lambda_3} + \frac{N_1^0 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}) e^{-\lambda_3 t} \right. \\ &\quad \left. + (\frac{N_2^0 \lambda_2}{\lambda_3 - \lambda_2} + \frac{N_1^0 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)}) e^{-\lambda_2 t} \right. \\ &\quad \left. + \frac{N_1^0 \lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} e^{-\lambda_1 t} \right] \\ &= v_3 [B_{33} e^{-\lambda_3 t} + B_{32} e^{-\lambda_2 t} + B_{31} e^{-\lambda_1 t}] \end{aligned}$$

Applying equation to three members, the transport equation for the first nuclide is as follows

$$\begin{aligned} \epsilon \frac{\partial N_1}{\partial t} + \epsilon V \frac{\partial N_1}{\partial z} - \epsilon D \frac{\partial^2 N_1}{\partial z^2} &= -R_{ad,1} - \epsilon \lambda_1 N_1 + \epsilon f_1 \\ (1-\epsilon) \frac{\partial N_1}{\partial t} &= R_{ad,1} - (1-\epsilon) \lambda_1 M_1 - k_1 M_1 (1-\epsilon) \\ (1-\epsilon) \frac{\partial L_1}{\partial t} &= -(1-\epsilon) \lambda_1 L_1 + k_1 M_1 (1-\epsilon) \\ M_1 &= k_{d1} N_1(z, t), \quad K_1 = 1 + \frac{1-\epsilon}{\epsilon} k_{d1} \end{aligned} \quad (4)$$

From equation (4)

$$K_1 \frac{\partial N_1}{\partial t} + \epsilon V \frac{\partial N_1}{\partial z} - \epsilon D \frac{\partial^2 N_1}{\partial z^2} + K_1 \lambda_1^* N_1 = f_1 \quad (5)$$

here let  $\frac{f_1(z, t)}{K_1} = f_1(t) \cdot \delta(z)$

$$\begin{aligned}
 N_1 &= \int_0^t \int_{-\infty}^{\infty} f_1(t-\theta) \cdot \delta(\xi) \frac{e^{-\lambda_1^* \theta}}{\sqrt{4\pi \kappa v_1 \theta}} \exp\left[-\frac{(z-\xi-v_1 \theta)^2}{4\kappa v_1 \theta}\right] d\xi d\theta \\
 &= \int_0^t f_1(t-\theta) \frac{e^{-\lambda_1^* \theta}}{\sqrt{4\pi \kappa v_1 \theta}} \exp\left[-\frac{(z-v_1 \theta)^2}{4\kappa v_1 \theta}\right] d\theta \\
 &= \int_0^t f_1(t-\theta) \cdot e^{-\lambda_1^* \theta} F(v_1 \theta, z-v_1 \theta) d\theta
 \end{aligned} \tag{6}$$

here  $F(v_1 \theta, z-v_1 \theta) = \frac{1}{\sqrt{4\pi \kappa v_1 \theta}} \exp\left[-\frac{(z-v_1 \theta)^2}{4\kappa v_1 \theta}\right]$

$$\kappa = \frac{D}{v}$$

From equation (6)

$$\begin{aligned}
 N_1 &= v_1 B_{11} e^{-\lambda_1 t} \int_0^t e^{-(\lambda_1^* - \lambda_1) \theta} F(v_1 \theta, z-v_1 \theta) d\theta \\
 &= B_{11} \cdot e^{-\lambda_1 t} e^{z/2\kappa} \frac{1}{2\sqrt{1+\frac{4\kappa(\lambda_1^* - \lambda_1)}{v_1}}} \left[ e^{-\frac{|z|}{2\kappa} \sqrt{1+\frac{4\kappa(\lambda_1^* - \lambda_1)}{v_1}}} \operatorname{erfc}\left(\frac{|z|-v_1 t}{\sqrt{4\kappa v_1 t}}\right) \right. \\
 &\quad \left. - e^{\frac{|z|}{2\kappa} \sqrt{1+\frac{4\kappa(\lambda_1^* - \lambda_1)}{v_1}}} \operatorname{erfc}\left(\frac{|z|+v_1 t}{\sqrt{4\kappa v_1 t}}\right) \right] \\
 &= \underline{\underline{B_{11} \cdot E(1,1; 1)}}
 \end{aligned} \tag{7}$$

here

$$\begin{aligned}
 E(i,j;k) &= \frac{e^{-\beta_{ij} t + z/2\kappa}}{2\sqrt{\gamma_{ijk}}} \left[ e^{-\frac{|z|}{2\kappa} \sqrt{\gamma_{ijk}}} \operatorname{erfc}\left(\frac{|z|-v_k t \sqrt{\gamma_{ijk}}}{\sqrt{4\kappa v_k t}}\right) \right. \\
 &\quad \left. - e^{\frac{|z|}{2\kappa} \sqrt{\gamma_{ijk}}} \operatorname{erfc}\left(\frac{|z|+v_k t \sqrt{\gamma_{ijk}}}{\sqrt{4\kappa v_k t}}\right) \right] \\
 &\quad (\gamma_{ijk} > 0)
 \end{aligned}$$

$$\gamma_{ijk} = 1 + 4\kappa(\lambda_k^* - \beta_{ij})/v_k \tag{8}$$

$$\Lambda_{kj} = \frac{\lambda_k^*}{v_k} - \frac{\lambda_j^*}{v_j}, \quad \Gamma_{kj} = \frac{1}{v_k} - \frac{1}{v_j}$$

$$\beta_{ij} = \begin{cases} \lambda_j & : i = j \\ \frac{\Lambda_{ij}}{\Gamma_{ij}} & ; i \neq j \end{cases}$$

For the second nuclide

$$\begin{aligned} \varepsilon \frac{\partial N_2}{\partial t} + \varepsilon V \frac{\partial N_2}{\partial z} - \varepsilon D \frac{\partial^2 N_2}{\partial z^2} &= -R_{ad,2} - \varepsilon \lambda_2 N_2 + \varepsilon \lambda_1 N_1 + \varepsilon f_2 \\ (1-\varepsilon) \frac{\partial M_2}{\partial t} &= R_{ad,2} - (1-\varepsilon) \lambda_2 M_2 + (1-\varepsilon) \lambda_1 M_1 - (1-\varepsilon) k_2 M_2 + (1-\varepsilon) S_1^{LM} \lambda_1 L_1 \\ (1-\varepsilon) \frac{\partial L_2}{\partial t} &= -(1-\varepsilon) \lambda_2 L_2 + (1-\varepsilon) S_1^{LL} \lambda_1 L_1 + (1-\varepsilon) k_2 M_2 \\ N_2 = M_2/k_{d2} &\quad , \quad 1 + \frac{1-\varepsilon}{\varepsilon} k_{d2} = K_2 \end{aligned} \quad (9)$$

From equation (9)

$$K_2 \frac{\partial N_2}{\partial t} + \varepsilon V \frac{\partial N_2}{\partial z} - \varepsilon D \frac{\partial^2 N_2}{\partial z^2} + K_2 \lambda_2^* N_2 = K_1 \lambda_1 N_1 + \frac{1-\varepsilon}{\varepsilon} S_1^{LM} \lambda_1 L_1 + f_2 \quad (10)$$

$$\begin{aligned} \therefore N_2(z, t) &= \int_0^t f_2(t-\theta_2) e^{-\lambda_2^* \theta_2} F(v_2 \theta_2, z-v_2 \theta_2) \\ &+ \int_0^t \int_{-\infty}^{\infty} \left\{ \frac{K_1}{K_2} \lambda_1 N_1(\xi, t-\theta_2) + \frac{1-\varepsilon}{\varepsilon K_2} S_1^{LM} \lambda_1 L_1(\xi, t-\theta_2) \right\} \\ &\times e^{-\lambda_2^* \theta_2} F(v_2 \theta_2, z-\xi-v_2 \theta_2) d\theta_2 \cdot d\xi \end{aligned} \quad (11)$$

Taking Laplace transform for equation (11)

$$\begin{aligned} \tilde{N}_2(z, s) &= \tilde{f}_2(s) \cdot L[e^{-\lambda_2^* t} F(v_2 t, z-v_2 t)] \\ &+ \int_{-\infty}^{\infty} \left[ \frac{K_1}{K_2} \lambda_1 \tilde{N}_1(\xi, s) + \frac{1-\varepsilon}{\varepsilon K_2} S_1^{LM} \tilde{L}_1(\xi, s) \right] d\xi \end{aligned} \quad (12)$$

here

$$\begin{aligned} \tilde{N}_1(\xi, s) &= \tilde{f}_1(s) \cdot L[e^{-\lambda_1^* \theta_1} F(v_1 \theta_1, \xi-v_1 \theta_1)] \\ L_1(\xi, t) &= \int_0^t K_1 K_{d1} N_1(\xi, \tau) e^{-\lambda_1(t-\tau)} d\tau \\ \tilde{L}_1(\xi, s) &= \frac{k_1 k_{d1} \tilde{f}_1(s)}{s + \lambda_1} \cdot L[e^{-\lambda_1^* t} F(v_1 t, \xi-v_1 t)] \end{aligned}$$

$$\begin{aligned}
\therefore N_2(z, s) = & \tilde{f}_2(s) L[e^{-\lambda_2^* \theta} F(v_2 \theta, z - v_2 \theta)] \\
& + \int_{-\infty}^{\infty} \frac{K_1 \lambda_1}{K_2} \tilde{f}_1(s) \cdot L_{\theta_1} [e^{-\lambda_1^* \theta_1} F(v_1 \theta_1, \xi_1 - v_1 \theta_1)] \\
& \times L_{\theta_2} [e^{-\lambda_2^* \theta_2} F(v_2 \theta_2, z - \xi_1 - v_2 \theta_2)] d\xi_1 \\
& + \int_{-\infty}^{\infty} \frac{(1-\varepsilon) s_1^{LM} \lambda_1 k_1 k d_1}{\varepsilon K_2 (s + \lambda_1)} \tilde{f}_1(s) L_{\theta_1} [e^{-\lambda_1^* \theta_1} F(v_1 \theta_1, \xi_1 - v_1 \theta_1)] \\
& \times L_{\theta_2} [e^{-\lambda_2^* \theta_2} F(v_2 \theta_2, z - \xi_1 - v_2 \theta_2)] d\xi_1
\end{aligned} \tag{13}$$

The first term is;

$$N_{1,2}(z, s) = \tilde{f}_2(s) \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \sqrt{1 + \frac{4\kappa(s + \lambda_2^*)}{v_2}}}{v_2 \sqrt{1 + \frac{4\kappa(s + \lambda_2^*)}{v_2}}} = \tilde{f}_2(s) \frac{1}{v_2} \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}{\delta_2} \tag{14a}$$

here

$$\delta_2 = \sqrt{1 + \frac{4\kappa(s + \lambda_2^*)}{v_2}}$$

generally

$$\delta_i = \sqrt{1 + \frac{4\kappa(s + \lambda_i^*)}{v_i}} \quad (i=1, 2, 3) \tag{14b}$$

$$\begin{aligned}
N_{2,1}(z, t) = & v_2 B_{22} \int_0^t \frac{1}{v_2} e^{-\lambda_2(t-\theta)} L^{-1} \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}{\delta_2} d\theta \\
& + v_2 B_{21} \int_0^t \frac{1}{v_2} e^{-\lambda_1(t-\theta)} L^{-1} \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}{\delta_2} d\theta \\
= & B_{22} E(2, 2; 2) + B_{21} E(1, 1; 2)
\end{aligned} \tag{15}$$

The second term is;

$$\begin{aligned}
& \int_{-\infty}^{\infty} L_{\theta_1} [e^{-\lambda_1^* \theta} F(v_1 \theta_1, \xi_1 - v_1 \theta_1)] L_{\theta_2} [e^{-\lambda_2^* \theta_2} F(v_2 \theta_2, z - \xi_1 - v_2 \theta_2)] d\xi_1 \\
= & L_{\theta_1} L_{\theta_2} [e^{-\lambda_1^* \theta_1 - \lambda_2^* \theta_2} F(v_1 \theta_1 + v_2 \theta_2, z - v_1 \theta_1 - v_2 \theta_2)] \\
= & \frac{\frac{1}{v_1 v_2} \cdot e^{\frac{z}{2\kappa}}}{(\lambda_2^* + s)} \left[ \frac{e}{\delta_1} - \frac{|z|}{2\kappa} \delta_1 - \frac{e}{\delta_2} - \frac{|z|}{2\kappa} \delta_2 \right] \tag{2} \\
& \tag{16a}
\end{aligned}$$

$$\begin{aligned}
N_{2,2}(z,s) &= \frac{K_1 \lambda_1}{K_2} \tilde{f}_1(s) \frac{1}{s+\beta_{12}} \frac{1}{v_1-v_2} \left[ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} - \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} \right] \\
&= \frac{K_1 \lambda_1 v_1 B_{11}}{K_2 (v_1-v_2)} \cdot \frac{1}{s+\lambda_1} \frac{1}{s+\beta_{12}} \left[ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} - \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} \right]
\end{aligned} \tag{16b}$$

$$\begin{aligned}
N_{2,2}(z,t) &= \frac{K_1 \lambda_1 v_1 B_{11}}{K_2 (v_1-v_2)} \cdot \frac{1}{(\beta_{12}-\lambda_1)} \left[ \left\{ \int_0^t e^{-\lambda_1(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} d\theta \right. \right. \\
&\quad - \left. \int_0^t e^{-\beta_{12}(t-\theta)} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} d\theta \right\} - \left\{ \int_0^t e^{-\lambda_1(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} d\theta \right. \\
&\quad \left. \left. - \int_0^t e^{-\beta_{12}(t-\theta)} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} d\theta \right\} \right] \\
&= \frac{K_1 \lambda_1 v_1 B_{11}}{K_2 (v_1-v_2) (\beta_{12}-\lambda_1)} [E(1,1;1)-E(1,2;1)-E(1,1;2)+E(1,2;2)]
\end{aligned} \tag{16c}$$

The third term is;

$$\begin{aligned}
N_{2,3}(z,s) &= \frac{(1-\epsilon) s_1^{LM} \lambda_1 k_1 k d_1}{\epsilon K_2 (v_1-v_2)} \frac{v_1 B_{11}}{(s+\lambda_1)^2 (s+\beta_{12})} \left[ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} - \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} \right] \\
&= \frac{(1-\epsilon) s_1^{LM} \lambda_1 k_1 k d_1 v_1 B_{11}}{\epsilon K_2 (v_1-v_2) (-\lambda_1+\beta_{12})^2} [e^{-\beta_{12}t} - \{1-(-\lambda_1+\beta_{12})t\} e^{-\lambda_1 t}] \\
&\quad \times \left[ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} - \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} \right]
\end{aligned} \tag{17}$$

$$\begin{aligned}
N_{2,3}(z,t) &= \frac{(1-\epsilon) s_1^{LM} \lambda_1 k_1 k d_1 v_1 B_{11}}{\epsilon K_2 (v_1-v_2) (-\lambda_1+\beta_{12})^2} \left[ \int_0^t e^{-\beta_{12}(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{\delta_1} d\theta \right. \\
&\quad - \left. \{1+(\lambda_1-\beta_{12})t\} \int_0^t e^{-\lambda_1(t-\theta)} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_1} d\theta \right. \\
&\quad - \left. \int_0^t e^{-\beta_{12}(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} d\theta \right. \\
&\quad + \left. \{1+(\lambda_1-\beta_{12})t\} \int_0^t e^{-\lambda_1(t-\theta)} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{\delta_2} d\theta \right] \\
&= \frac{(1-\epsilon) s_1^{LM} \lambda_1 k_1 k d_1 v_1 B_{11}}{\epsilon K_2 (v_1-v_2) (-\lambda_1+\beta_{12})^2} \{ \{ E(1,2;1)-E(1,2;2) \} \\
&\quad - \{1+(\lambda_1-\beta_{12})t\} \{ E(1,1;1)-E(1,1;2) \} \}
\end{aligned} \tag{18}$$

$$\text{here } \lambda_1^* = \lambda_1 + \frac{k_1(1-\varepsilon)kd_1}{\varepsilon K_1} \\ \therefore N_{2,3}(z,t) = \frac{B_{11} s_1^{LM} \lambda_1 (\lambda_1^* - \lambda_1) v_2}{(-\lambda_1 + \beta_{12})^2 (v_1 - v_2)} [ \{E(1,2;1) - E(1,2;2)\} \\ - \{1 + (\lambda_1 - \beta_{12})t\} \{E(1,1;1) - E(1,1;2)\}] \quad (19)$$

$$\begin{aligned}
 N_2(z, t) = & \frac{B_{22}E(2, 2; 2) + B_{21}E(1, 1; 2)}{1} \\
 & + \frac{B_{11}k_1\lambda_1 v_1}{K_2(v_1 - v_2)(\beta_{12} - \lambda_1)} [E(1, 1; 1) - E(1, 2; 1) - E(1, 1; 2) + E(1, 2; 2)] \\
 & + \frac{B_{11}s_1^{LM} \lambda_1 (\lambda_1^* - \lambda_1) v_2}{(-\lambda_1 + \beta_{12})^2 (v_1 - v_2)} [\{E(1, 2; 1) - E(1, 2; 2)\}] \\
 & - \{1 + (\lambda_1 - \beta_{12})t\} \{E(1, 1; 1) - E(1, 1; 2)\} \quad (20)
 \end{aligned}$$

For the third nuclide

$$\begin{aligned} \epsilon \frac{\partial N_3}{\partial t} + \epsilon V \frac{\partial N_3}{\partial z} - \epsilon D \frac{\partial^2 N}{\partial z^2} &= -R_{ad,3} - \epsilon \lambda_3 N_3 + \epsilon \lambda_2 N_2 + \epsilon f_3 \\ (1-\epsilon) \frac{\partial M_3}{\partial t} &= R_{ad,3} - (1-\epsilon) \lambda_3 M_3 + (1-\epsilon) \lambda_2 M_2 - (1-\epsilon) k_3 M_3 + (1-\epsilon) s_2^{LM} \lambda_2 L_2 \\ (1-\epsilon) \frac{\partial L_3}{\partial t} &= -(1-\epsilon) \lambda_3 L_3 + (1-\epsilon) s_2^{LL} \lambda_2 L_2 + (1-\epsilon) k_3 M_3 \end{aligned} \quad (21)$$

From equation (21)

$$K_3 \frac{\partial N_3}{\partial z} + \epsilon V \frac{\partial N_3}{\partial z} - \epsilon D \frac{\partial^2 N_3}{\partial z^2} + K_3 \lambda_3^* N_3 = K_2 \lambda_2 N_2 + \frac{1-\epsilon}{\epsilon} s_2^{LM} \lambda_2 L_2 + f_3 \quad (22)$$

$$\begin{aligned} \therefore N_3(z, t) &= \int_0^t f_3(t-\theta_3) e^{-\lambda_3^* \theta_3} F(v_3 \theta_3, z - v_3 \theta_3) d\theta_3 \\ &+ \int_0^t \int_{-\infty}^{\infty} \left\{ \frac{k_2}{K_3} \lambda_2 N_2(\xi, t-\theta_3) + \frac{1-\varepsilon}{\varepsilon K_3} s_2^{LM} \lambda_2 L_2(\xi, t-\theta_3) \right\} e^{-\lambda_3^* \theta_3} \\ &\quad \times F(v_3 \theta_3, z - \xi - v_3 \theta_3) d\theta_3 \cdot d\xi \end{aligned} \quad (23)$$

Taking Laplace transform for equation (23)

$$\begin{aligned} \tilde{N}_3(z, s) &= \tilde{f}_3(s) \int_L [e^{-\lambda_3^* t} F(v_3 t, z - v_3 t)] \\ &\quad + \int_{-\infty}^{\infty} \left[ \frac{K_2 \lambda_2}{K_3} \tilde{N}_2(\xi_2, s) + \frac{1-\epsilon}{\epsilon K_3} s_2^{LM} \lambda_2 \tilde{L}_2(\xi_2, s) \right] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_2 \end{aligned} \quad (24)$$

Here  $\tilde{N}_2(\xi_2, s)$  is expressed by equation (12)

From equation (9)

$$\frac{\partial L_2}{\partial t} + \lambda_2 L_2 = \lambda_1 s_1^{LL} L_1 + k_2 k d_2 N_2$$

$$\therefore L_2(z, t) = \int_0^t e^{-\lambda_2(t-\theta)} [\lambda_1 s_1^{LL} L_1(z, \theta_2) + k_2 k d_2 N_2(z, \theta_2)] d\theta_2 \quad (25)$$

$$\tilde{L}_2(z, s) = \frac{1}{s + \lambda_2} \{ \lambda_1 s_1^{LL} \tilde{L}_1(z, s) + k_2 k d_2 \tilde{N}_2(z, s) \} \quad (26)$$

$$\tilde{N}_3(z, s) = \tilde{f}_3(s) \int_L [e^{-\lambda_3^* t} F(v_3 t, z - v_3 t)]$$

$$\begin{aligned} &\left[ \int_{-\infty}^{\infty} \tilde{f}_2(s) \int_L [e^{-\lambda_2^* t} F(v_2 t, \xi_2 - v_2 t)] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_2 \right. \\ &+ \frac{K_2}{K_3} \lambda_2 \times \left. \begin{aligned} &+ \int_{-\infty}^{\infty} \frac{K_1 \lambda_1}{K_2} \tilde{f}_1(s) \int_L [e^{-\lambda_1^* t} F(v_1 t, \xi_1 - v_1 t)] \int_L [e^{-\lambda_2^* t} F(v_2 t, \xi_2 - \xi_1 - v_2 t)] \\ &\times \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_1 d\xi_2 \\ &+ \int_{-\infty}^{\infty} \frac{1-\epsilon}{\epsilon K_2} s_1^{LM} \frac{\lambda_1 k_1 k d_1}{s + \lambda_1} \tilde{f}_1(s) \int_L [e^{-\lambda_1^* t} F(v_1 t, \xi_1 - v_1 t)] \\ &\times \int_L [e^{-\lambda_2^* t} F(v_2 t, \xi_2 - \xi_1 - v_2 t)] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_1 d\xi_2 \\ &\left. \begin{aligned} &\int_{-\infty}^{\infty} \frac{1}{s + \lambda_2} \lambda_1 s_1^{LL} \frac{k_1 k d_1}{s + \lambda_1} \tilde{f}_1(s) \int_L [e^{-\lambda_1^* t} F(v_1 t, \xi_2 - v_1 t)] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_2 \\ &+ \int_{-\infty}^{\infty} \frac{1}{s + \lambda_2} k_2 k d_2 \tilde{f}_2(s) \int_L [e^{-\lambda_2^* t} F(v_2 t, \xi_2 - v_2 t)] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_2 \\ &+ \int_{-\infty}^{\infty} \frac{1}{s + \lambda_2} k_2 k d_2 \frac{K_1 \lambda_1}{K_2} \tilde{f}_1(s) \int_L [e^{-\lambda_1^* t} F(v_1 t, \xi_1 - v_1 t)] \\ &\times \int_L [e^{-\lambda_2^* t} F(v_2 t, \xi_2 - \xi_1 - v_2 t)] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_1 d\xi_2 \\ &+ \int_{-\infty}^{\infty} \frac{1}{s + \lambda_2} k_2 k d_2 \frac{1-\epsilon}{\epsilon K_2} s_1^{LM} \frac{\lambda_1 k_1 k d_1}{s + \lambda_1} \tilde{f}_1(s) \int_L [e^{-\lambda_1^* t} F(v_1 t, \xi_1 - v_1 t)] \\ &\times \int_L [e^{-\lambda_2^* t} F(v_2 t, \xi_2 - \xi_1 - v_2 t)] \int_L [e^{-\lambda_3^* t} F(v_3 t, z - \xi_2 - v_3 t)] d\xi_1 d\xi_2 \end{aligned} \right] \end{aligned} \quad (27)$$

here let

$$\begin{aligned} \tilde{N}_{3,1}(z,s) &= \tilde{f}_3(s) L[e^{-\lambda_3^* t} F(v_3 t, z - v_3 t)] \\ \tilde{N}_{3,2}(z,s) &= \int_{-\infty}^{\infty} \frac{K_2}{K_3} \lambda_2 \cdot \tilde{f}_2(s) L[\quad] L[\quad] d\xi_2 \\ \tilde{N}_{3,3}(z,s) &= \iint_{-\infty}^{\infty} \frac{K_2}{K_3} \lambda_2 \frac{K_1 \lambda_1}{K_2} \tilde{f}_1(s) L[\quad] L[\quad] L[\quad] d\xi_1 d\xi_2 \\ &\vdots \\ &\vdots \\ \tilde{N}_{3,8}(z,s) &= \iint_{-\infty}^{\infty} \frac{1}{s + \lambda_2} k_2 k d_2 \frac{1 - \epsilon}{\epsilon K_2} s_1^{LM} \frac{\lambda_1 k_1 k d_1}{s + \lambda_1} \tilde{f}_1(s) L[\quad] L[\quad] L[\quad] d\xi_1 d\xi_2 \end{aligned} \quad (28)$$

$$\tilde{N}_{3,1}(z,s) = \tilde{f}_3(s) \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \sqrt{1 + \frac{4\kappa(s + \lambda_3^*)}{v_3}}}{v_3 \sqrt{1 + \frac{4\kappa(s + \lambda_3^*)}{v_3}}} = \tilde{f}_3(s) \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{v_3 \delta_3} \quad (29)$$

$$\begin{aligned} N_{3,1}(z,t) &= B_{33} \int_0^t e^{-\lambda_3(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{\delta_3} d\theta \\ &+ B_{32} \int_0^t e^{-\lambda_2(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{\delta_3} d\theta \\ &+ B_{31} \int_0^t e^{-\lambda_1(t-\theta)} L^{-1} \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{\delta_3} d\theta \end{aligned}$$

$$= B_{33} E(3,3;3) + B_{32} E(2,2;3) + B_{31} E(1,1;3) \quad (30)$$

$$\tilde{N}_{3,2}(z,s) = \frac{K_2}{K_3} \lambda_2 \tilde{f}_2(s) \frac{1}{s + \beta_{23}} \frac{1}{v_2 - v_3} \left\{ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_2}{\delta_2} - \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{\delta_3} \right\} \quad (31)$$

$$\begin{aligned} \therefore N_{3,2}(z,t) &= \frac{k_2 \lambda_2 v_2}{K_3 (v_2 - v_3)} \frac{B_{22}}{\beta_{23} - \lambda_2} \{ E(2,2;2) - E(2,2;3) - E(2,3;2) + E(2,3;3) \} \\ &+ \frac{K_2 \lambda_2 v_2}{K_3 (v_2 - v_3)} \frac{B_{21}}{\beta_{23} - \lambda_1} \{ E(1,1;2) - E(1,1;3) - E(2,3;2) + E(2,3;3) \} \end{aligned} \quad (32)$$

$$N_{3,3}(z,s) = \frac{K_1 \lambda_1 \lambda_2}{K_3} \tilde{f}_1(s) \frac{1}{v_1 v_2 v_3} \left\{ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{g_{13} g_{23} \delta_3} + \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_2}{g_{32} g_{12} \delta_2} + \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_1}{g_{21} g_{31} \delta_1} \right\} \quad (33)$$

$$\text{here } g_{ij} = \frac{s+\lambda_i^*}{v_i} - \frac{s+\lambda_j^*}{v_j}$$

$$\begin{aligned}\therefore N_{3,3}(z,t) &= -\frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{13} \Gamma_{23}} \cdot \frac{1}{(-\lambda_1 + \beta_{13})(-\beta_{23} + \lambda_1)} E(1,1;3) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{13} \Gamma_{23}} \cdot \frac{1}{(-\lambda_1 + \beta_{13})(-\beta_{13} + \beta_{23})} E(1,3;3) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{13} \Gamma_{23}} \cdot \frac{1}{(-\beta_{13} + \beta_{23})(-\beta_{23} + \lambda_1)} E(2,3;3) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{32} \Gamma_{12}} \cdot \frac{1}{(-\lambda_1 + \beta_{32})(-\beta_{12} + \lambda_1)} E(1,1;2) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{32} \Gamma_{12}} \cdot \frac{1}{(-\lambda_1 + \beta_{32})(-\beta_{32} + \beta_{12})} E(2,3;2) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{32} \Gamma_{12}} \cdot \frac{1}{(-\beta_{32} + \beta_{12})(-\beta_{12} + \lambda_1)} E(1,2;2) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{21} \Gamma_{31}} \cdot \frac{1}{(-\lambda_1 + \beta_{21})(\beta_{31} + \lambda_1)} E(1,1;1) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{21} \Gamma_{31}} \cdot \frac{1}{(-\lambda_1 + \beta_{21})(-\beta_{21} + \beta_{31})} E(1,2;1) \\ &\quad - \frac{\lambda_1 \lambda_2 B_{11}}{v_1 v_2 \Gamma_{21} \Gamma_{31}} \cdot \frac{1}{(\beta_{21} + \beta_{31})(-\beta_{31} + \lambda_1)} E(1,3;1)\end{aligned}$$

$$\begin{aligned}\tilde{N}_{3,4}(z,s) &= \frac{\lambda_2(1-\epsilon)}{\epsilon \kappa_3} s_1^{\text{LM}} \lambda_1 k_1 k d_1 \frac{\tilde{f}_1(s)}{s+\lambda_1} \frac{1}{v_1 v_2 v_3} \left\{ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{g_{13} g_{23} \delta_3} + \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_2}{g_{32} g_{12} \delta_2} \right. \\ &\quad \left. + \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_1}{g_{21} g_{31} \delta_1} \right\} \quad (35)\end{aligned}$$

$$\begin{aligned}\therefore N_{3,4}(z,t) &= \frac{s_1^{\text{LM}} \lambda_1 \lambda_2 (\lambda_1^* - \lambda_1) B_{11}}{v_1 v_2 \Gamma_{13} \Gamma_{23}} \\ &\quad \times \left[ \begin{aligned} &\left\{ \frac{1}{(-\lambda_1 + \beta_{13})(-\lambda_1 + \beta_{23})} t - \frac{(-\lambda_1 + \beta_{13}) + (-\lambda_1 + \beta_{23})}{(-\lambda_1 + \beta_{13})^2 (-\lambda_1 + \beta_{23})^2} \right\} E(1,1;3) \\ &+ \frac{1}{(-\beta_{13} + \lambda_1)^2 (-\beta_{13} + \beta_{23})} E(1,3;3) \\ &+ \frac{1}{(-\beta_{23} + \lambda_1)^2 (-\beta_{23} + \beta_{13})} E(2,3;3) \end{aligned} \right]\end{aligned}$$

$$\begin{aligned}
& + \frac{s_1^{\text{LM}} \lambda_1 \lambda_2 (\lambda_1^* - \lambda_1) B_{11}}{v_1 v_2 \Gamma_{32} \Gamma_{12}} \\
& \times \left[ \begin{array}{l} \frac{1}{(-\lambda_1 + \beta_{13})(-\lambda_1 + \beta_{12})} t - \frac{(-\lambda_1 + \beta_{32}) + (-\lambda_1 + \beta_{12})}{(-\lambda_1 + \beta_{32})^2 (-\lambda_1 + \beta_{12})^2} E(1,1;2) \\ + \frac{1}{(-\beta_{32} + \lambda_1)^2 (-\beta_{32} + \beta_{12})} E(2,3;2) \\ + \frac{1}{(-\beta_{12} + \lambda_1)^2 (-\beta_{12} + \beta_{32})} E(1,2;2) \end{array} \right] \\
& + \frac{s_1^{\text{LM}} \lambda_1 \lambda_2 (\lambda_1^* - \lambda_1) B_{11}}{v_1 v_2 \Gamma_{21} \Gamma_{31}} \\
& \times \left[ \begin{array}{l} \frac{1}{(-\lambda_1 + \beta_{21})(-\lambda_1 + \beta_{31})} t - \frac{(-\lambda_1 + \beta_{21}) + (-\lambda_1 + \beta_{31})}{(-\lambda_1 + \beta_{21})^2 (-\lambda_1 + \beta_{31})^2} E(1,1;1) \\ + \frac{1}{(-\beta_{21} + \lambda_1)^2 (-\beta_{21} + \beta_{31})} E(1,2;1) \\ + \frac{1}{(-\beta_{31} + \lambda_1)^2 (-\beta_{31} + \beta_{21})} E(1,3;1) \end{array} \right] \tag{36}
\end{aligned}$$

$$\begin{aligned}
\tilde{N}_{3,5}(z, s) &= \frac{1-\epsilon}{\epsilon K_3} s_2^{\text{LM}} \lambda_1 \lambda_2 s_1^{\text{LL}} k_1 k d_1 \frac{1}{s+\lambda_1} \frac{1}{s+\lambda_2} \frac{1}{s+\beta_{31}} \tilde{f}_1(s) \frac{1}{v_1-v_3} \\
&\times \left\{ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_1}{\delta_1} - \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa}} \delta_3}{\delta_3} \right\} \tag{37}
\end{aligned}$$

$$\begin{aligned}
N_{3,5}(z, t) &= \frac{s_1^{\text{LM}} s_1^{\text{LL}} \lambda_1 \lambda_2 (\lambda_1^* - \lambda_1) v_3 B_{11}}{v_1 - v_3} \\
&\times \left[ \begin{array}{l} \frac{1}{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{31})} t - \frac{(-\lambda_1 + \lambda_2) + (-\lambda_1 + \beta_{31})}{(-\lambda_1 + \lambda_2)^2 (-\lambda_1 + \beta_{31})^2} \times \{E(1,1;1) - E(1,1;3)\} \\ + \frac{1}{(-\lambda_2 + \lambda_1)^2 (-\lambda_2 + \beta_{31})} \{E(2,2;1) - E(2,2;3)\} \\ + \frac{1}{(-\beta_{31} + \lambda_1)^2 (-\beta_{31} + \lambda_2)} \{E(1,3;1) - E(1,3;3)\} \end{array} \right] \tag{38}
\end{aligned}$$

$$\begin{aligned} \tilde{N}_{3,6}(z,s) &= \frac{1-\varepsilon}{\varepsilon K_3} s_2^{LM} \lambda_2 k_2 k d_2 \frac{1}{s+\lambda_2} \cdot \frac{1}{s+\beta_{23}} \tilde{f}_2(s) \cdot \frac{1}{v_2-v_3} \\ &\times \left\{ \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}{\delta_2} - \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_3}{\delta_3} \right\} \end{aligned} \quad (39)$$

$$\begin{aligned} \therefore N_{3,6}(z,t) &= \frac{s_2^{LM} \lambda_2 (\lambda_2^* - \lambda_2) v_3}{v_2 - v_3} B_{22} \frac{1}{(-\lambda_2 + \beta_{23})^2} \left[ \begin{array}{l} \{E(2,3;2) - E(2,3;3)\} \\ - \{1 + (\lambda_2 - \beta_{23})t\} \times \{E(2,2;2) - E(2,2;3)\} \end{array} \right] \\ &- \frac{s_2^{LM} \lambda_2 (\lambda_2^* - \lambda_2) v_3}{v_2 - v_3} B_{21} \left[ \begin{array}{l} \frac{1}{(-\lambda_2 + \lambda_1)(-\beta_{23} + \lambda_2)} \{E(2,2;2) - E(2,2;3)\} \\ + \frac{1}{(-\lambda_2 + \lambda_1)(-\lambda_1 + \beta_{23})} \{E(1,1;2) - E(1,1;3)\} \\ + \frac{1}{(-\lambda_1 + \beta_{23})(-\beta_{23} + \lambda_2)} \{E(2,3;2) - E(2,3;3)\} \end{array} \right] \end{aligned} \quad (40)$$

$$\begin{aligned} \tilde{N}_{3,7}(z,s) &= \frac{1-\varepsilon}{\varepsilon K_3} s_2 \lambda_2 k_2 k d_2 \frac{k_1 \lambda_1}{K_2} \frac{1}{s+\lambda_2} \tilde{f}_1(s) \frac{1}{v_1 v_2 v_3} \left\{ \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_3}{g_{13} g_{23} \delta_3} \right. \\ &\left. + \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}{g_{32} g_{12} \delta_2} + \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}{g_{21} g_{31} \delta_1} \right\} \end{aligned} \quad (41)$$

$$\begin{aligned} \therefore N_{3,7}(z,t) &= \frac{s_2^{LM} \lambda_1 \lambda_2 (\lambda_2^* - \lambda_2) B_{11}}{v_1 v_2 \Gamma_{13} \Gamma_{23}} \times \left[ \begin{array}{l} \frac{1}{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{13})(-\lambda_1 + \beta_{23})} E(1,1;3) \\ + \frac{1}{(-\lambda_2 + \lambda_1)(-\lambda_2 + \beta_{13})(-\lambda_2 + \beta_{23})} E(2,2;3) \\ + \frac{1}{(-\beta_{13} + \lambda_1)(-\beta_{13} + \lambda_2)(-\beta_{13} + \beta_{23})} E(1,3;3) \\ + \frac{1}{(-\beta_{23} + \lambda_1)(-\beta_{23} + \lambda_2)(-\beta_{23} + \beta_{13})} E(2,3;3) \end{array} \right] \end{aligned}$$

(to be continued)

$$\begin{aligned}
& + \frac{s_2^{\text{LM}} \lambda_1 \lambda_2 (\lambda_2^* - \lambda_2) B_{11}}{v_1 v_2 \Gamma_{32} \Gamma_{12}} \times \left[ \begin{array}{l} \frac{1}{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{32})(-\lambda_1 + \beta_{12})} E(1,1;2) \\ + \frac{1}{(-\lambda_2 + \lambda_1)(-\lambda_2 + \beta_{32})(-\lambda_2 + \beta_{12})} E(2,2;2) \\ + \frac{1}{(-\beta_{32} + \lambda_1)(-\beta_{32} + \lambda_2)(-\beta_{32} + \beta_{12})} E(2,3;2) \\ + \frac{1}{(-\beta_{12} + \lambda_1)(-\beta_{12} + \lambda_2)(-\beta_{12} + \beta_{32})} E(1,2;2) \end{array} \right] \\
& + \frac{s_2^{\text{LM}} \lambda_1 \lambda_2 (\lambda_2^* - \lambda_2) B_{11}}{v_1 v_2 \Gamma_{21} \Gamma_{31}} \times \left[ \begin{array}{l} \frac{1}{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{21})(-\lambda_1 + \beta_{31})} E(1,1;1) \\ + \frac{1}{(-\lambda_2 + \lambda_1)(-\lambda_2 + \beta_{21})(-\lambda_2 + \beta_{31})} E(2,2;1) \\ + \frac{1}{(-\beta_{21} + \lambda_1)(-\beta_{21} + \lambda_2)(-\beta_{21} + \beta_{31})} E(1,2;1) \\ + \frac{1}{(-\beta_{31} + \lambda_1)(-\beta_{31} + \lambda_2)(-\beta_{31} + \beta_{21})} E(1,3;1) \end{array} \right]
\end{aligned} \tag{42}$$

$$\begin{aligned}
N_{3,8}(z,s) &= \frac{1-\epsilon}{\epsilon K_3} s_2^{\text{LM}} \lambda_2 k_2 k d_2 \frac{k_1 k d_1}{\epsilon K_2} s_1^{\text{LM}} \frac{\lambda_1 (1-\epsilon) \tilde{f}_1(s)}{(s+\lambda_1)(s+\lambda_2)} \frac{1}{v_1 v_2 v_3} \\
&\times \left\{ \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_3}}{g_{13} g_{23} \delta_3} + \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_2}}{g_{32} g_{12} \delta_2} + \frac{e^{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_1}}{g_{21} g_{31} \delta_1} \right\}
\end{aligned} \tag{43}$$

$$\begin{aligned}
N_{3,8}(z,t) &= \frac{s_1^{\text{LM}} s_2^{\text{LM}} \lambda_1 \lambda_2 (\lambda_1 - \lambda_1^*)(\lambda_2 - \lambda_2^*) B_{11}}{v_1 v_2} \\
&\times \left[ \begin{array}{l} \frac{1}{\Gamma_{13} \Gamma_{23}} \cdot \left\{ \frac{1}{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{13})(-\lambda_1 + \beta_{23})} t \right. \\ \left. - \frac{(-\lambda_1 + \lambda_2)(-\lambda_1 + \beta_{13}) + (-\lambda_1 + \beta_{13})(-\lambda_1 + \beta_{23}) + (-\lambda_1 + \beta_{23})(-\lambda_1 + \lambda_2)}{(-\lambda_1 + \lambda_2)^2 (-\lambda_1 + \beta_{13})^2 (-\lambda_1 + \beta_{23})^2} \right\} E(1,1;3) \\ + \frac{1}{\Gamma_{13} \Gamma_{23}} \cdot \frac{1}{(-\lambda_2 + \lambda_1)^2 (-\lambda_2 + \beta_{13})(-\lambda_2 + \beta_{23})} E(2,2;3) \\ + \frac{1}{\Gamma_{13} \Gamma_{23}} \cdot \frac{1}{(-\beta_{13} + \lambda_1)^2 (-\beta_{13} + \lambda_2)(-\beta_{13} + \beta_{23})} E(1,3;3) \end{array} \right]
\end{aligned}$$

(to be continued)

$$\begin{aligned}
& + \frac{1}{\Gamma_{13}\Gamma_{23}} \cdot \frac{1}{(-\beta_{23}+\lambda_1)^2(-\beta_{23}+\lambda_2)(-\beta_{23}+\beta_{13})} E(2,3;3) \\
& + \frac{1}{\Gamma_{32}\Gamma_{12}} \left\{ \frac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{32})(-\lambda_1+\beta_{12})} t \right. \\
& \quad \left. - \frac{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{32})+(-\lambda_1+\beta_{32})(-\lambda_1+\beta_{12})+(-\lambda_1+\beta_{12})(-\lambda_1+\lambda_2)}{(-\lambda_1+\lambda_2)^2(-\lambda_1+\beta_{32})^2(-\lambda_1+\beta_{12})^2} \right\} E(1,1;2) \\
& + \frac{1}{\Gamma_{32}\Gamma_{12}} \cdot \frac{1}{(-\lambda_2+\lambda_1)^2(-\lambda_2+\beta_{32})(-\lambda_2+\beta_{12})} E(2,2;2) \\
& + \frac{1}{\Gamma_{32}\Gamma_{12}} \cdot \frac{1}{(-\beta_{32}+\lambda_1)^2(-\beta_{32}+\lambda_2)(-\beta_{32}+\beta_{12})} E(2,3;2) \\
& + \frac{1}{\Gamma_{32}\Gamma_{12}} \cdot \frac{1}{(-\beta_{12}+\lambda_1)^2(-\beta_{12}+\lambda_2)(-\beta_{12}+\beta_{32})} E(1,2;2) \\
& + \frac{1}{\Gamma_{21}\Gamma_{31}} \left\{ \frac{1}{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{21})(-\lambda_1+\beta_{31})} t \right. \\
& \quad \left. - \frac{(-\lambda_1+\lambda_2)(-\lambda_1+\beta_{21})+(-\lambda_1+\beta_{21})(-\lambda_1+\beta_{31})+(-\lambda_1+\beta_{31})(-\lambda_1+\lambda_2)}{(-\lambda_1+\lambda_2)^2(-\lambda_1+\beta_{21})^2(-\lambda_1+\beta_{31})^2} \right\} E(1,1;1) \\
& + \frac{1}{\Gamma_{21}\Gamma_{31}} \cdot \frac{1}{(-\lambda_2+\lambda_1)^2(-\lambda_2+\beta_{21})(-\lambda_2+\beta_{31})} E(2,2;1) \\
& + \frac{1}{\Gamma_{21}\Gamma_{31}} \cdot \frac{1}{(-\beta_{21}+\lambda_1)^2(-\beta_{21}+\lambda_2)(-\beta_{21}+\beta_{31})} E(1,2;1) \\
& + \frac{1}{\Gamma_{21}\Gamma_{31}} \cdot \frac{1}{(-\beta_{31}+\lambda_1)(-\beta_{31}+\lambda_2)(-\beta_{31}+\beta_{21})} E(1,3;1)
\end{aligned} \tag{44}$$

Here  $\tilde{N}_3(z, s) = \sum_{i=1}^8 N_{3,i}(z, s)$

$$\tilde{N}_3(z, t) = \sum_{i=1}^8 N_{3,i}(z, t) \tag{45}$$

### 3. Numerical Demonstration

As the detailed mechanism and the rate of mineralization are not known well, it is difficult to evaluate the effect quantitatively. So we made a parametric study to understand the general effect of mineralization on migration pattern.

Calculation was carried out with the help of computer code MINELO1<sup>(\*\*\*)</sup> developed for the transport analysis with reversible mineralization.

From equation (2), we know that the effect of mineralization is expressed with the increase of  $\lambda_i^*$  value as follows

$$\begin{aligned}\lambda_i^* &= \lambda_i + \frac{k_i(1-\varepsilon)kd_i}{\varepsilon K_i} \\ &= \lambda_i + k_i(1 - \frac{1}{K_i})\end{aligned}\tag{46}$$

Numerical demonstration has been carried out in the case of  $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$  decay chain.

The nuclear data and other parameters used are shown in Table 1. Figure 2 and Fig. 3 are the migration profiles in the case of irreversible mineralization reaction. Irreversible mineralization means that daughter nuclide from the mineralized precursor by decay is fixed in the mineral.

Figure 2 shows the effect of precursors' mineralization to the concentration profile of  $^{226}\text{Ra}$  in the case of transient equilibrium sources at  $t = 0$ . Figure 3 is the profile for initial no daughter source at  $t = 0$ . Here  $N_1^0$  represents the concentration of  $^{234}\text{U}$  at  $z = 0$  just after the beginning of leaching. As shown in these figures the mineralization of  $^{230}\text{Th}$  has a marked effect on the  $^{226}\text{Ra}$  concentration profile. Figure 4 shows the effect of irreversible mineralization for  $^{234}\text{U}$  itself. Figure 5 shows the comparison of the reversible and irreversible mineralization of  $^{230}\text{Th}$  in  $^{234}\text{U} \rightarrow ^{230}\text{Th}$  decay chain. In this figure  $k_1^{\text{LM}} = 0$  means the irreversible mineralization and  $k_1^{\text{LM}} = 1$  means the reversible one.

Figure 6 is the  $^{226}\text{Ra}$  concentration profile in the case of  $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$  decay chain. It is an interesting phenomenon that the concentration of reversible mineralization exceed the one of non-mineralization in the range of  $z < 1.7 \times 10^{-2}$  km and  $z > 1.5 \times 10^{-1}$  km. This is due to the delay of release of  $^{226}\text{Ra}$  by the reversible mineralization of  $^{234}\text{U}$  and  $^{230}\text{Th}$ .

This is an important phenomena for evaluating the ingestion hazard of waste.

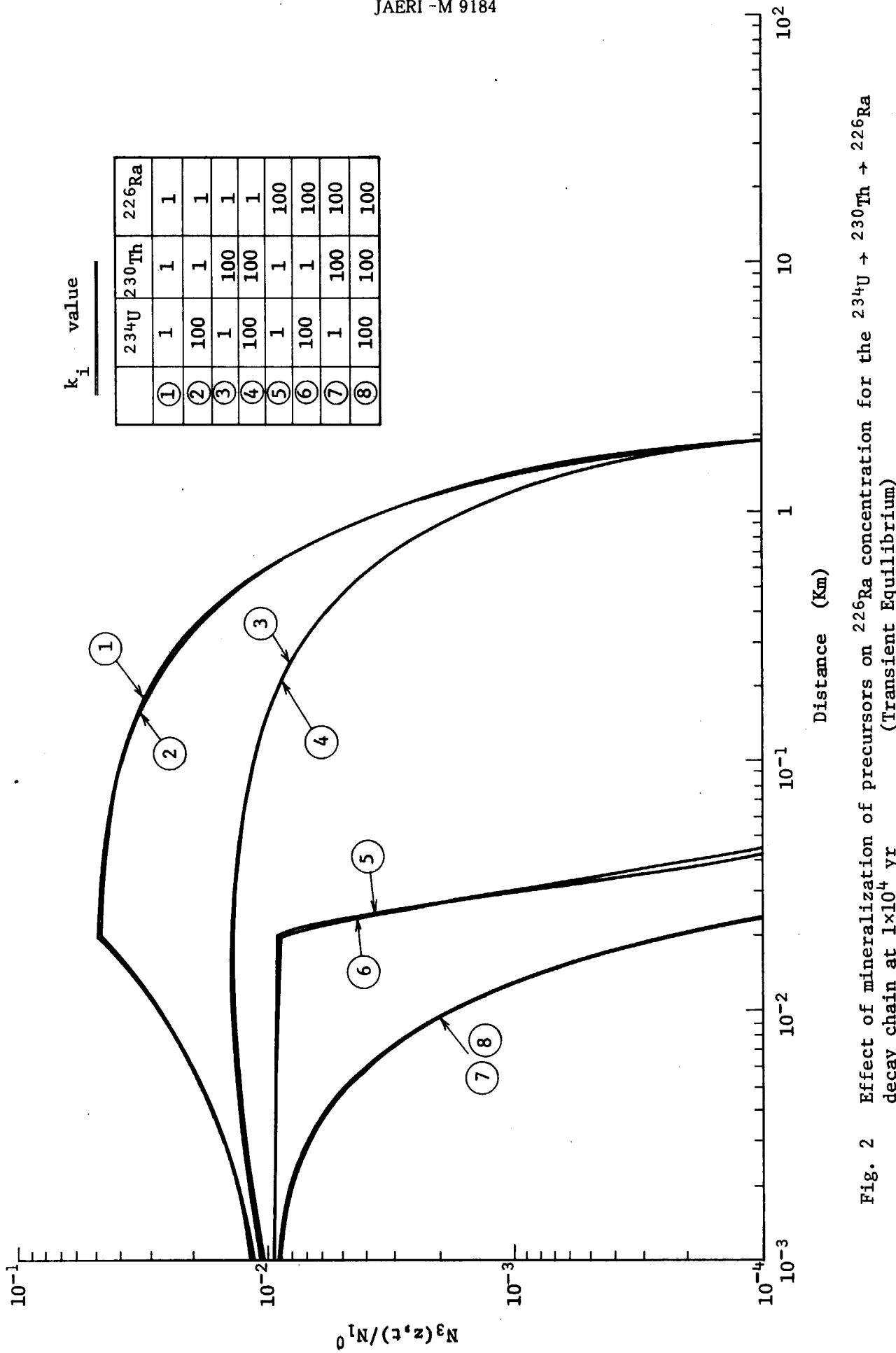
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(\*\*\*) Appendix [3]

Table 1 Nuclear Data and Other Parameters

	$^{234}\text{U}$	$^{230}\text{Th}$	$^{226}\text{Ra}$
Half life (yr)	$2.44 \times 10^5$	$7.7 \times 10^4$	$1.6 \times 10^3$
Decay constant $\lambda_1$ (1/yr)	$2.84 \times 10^{-6}$	$9.00 \times 10^{-6}$	$4.33 \times 10^{-4}$
Overall sorption coefficient $K_1$ ( - )	$1 \times 10^4$	$5 \times 10^4$	$5 \times 10^2$

Velocity of water       $V = 100$  m/yrDispersion coefficient  $D = 1 \times 10^{-1}$  m<sup>2</sup>/yr



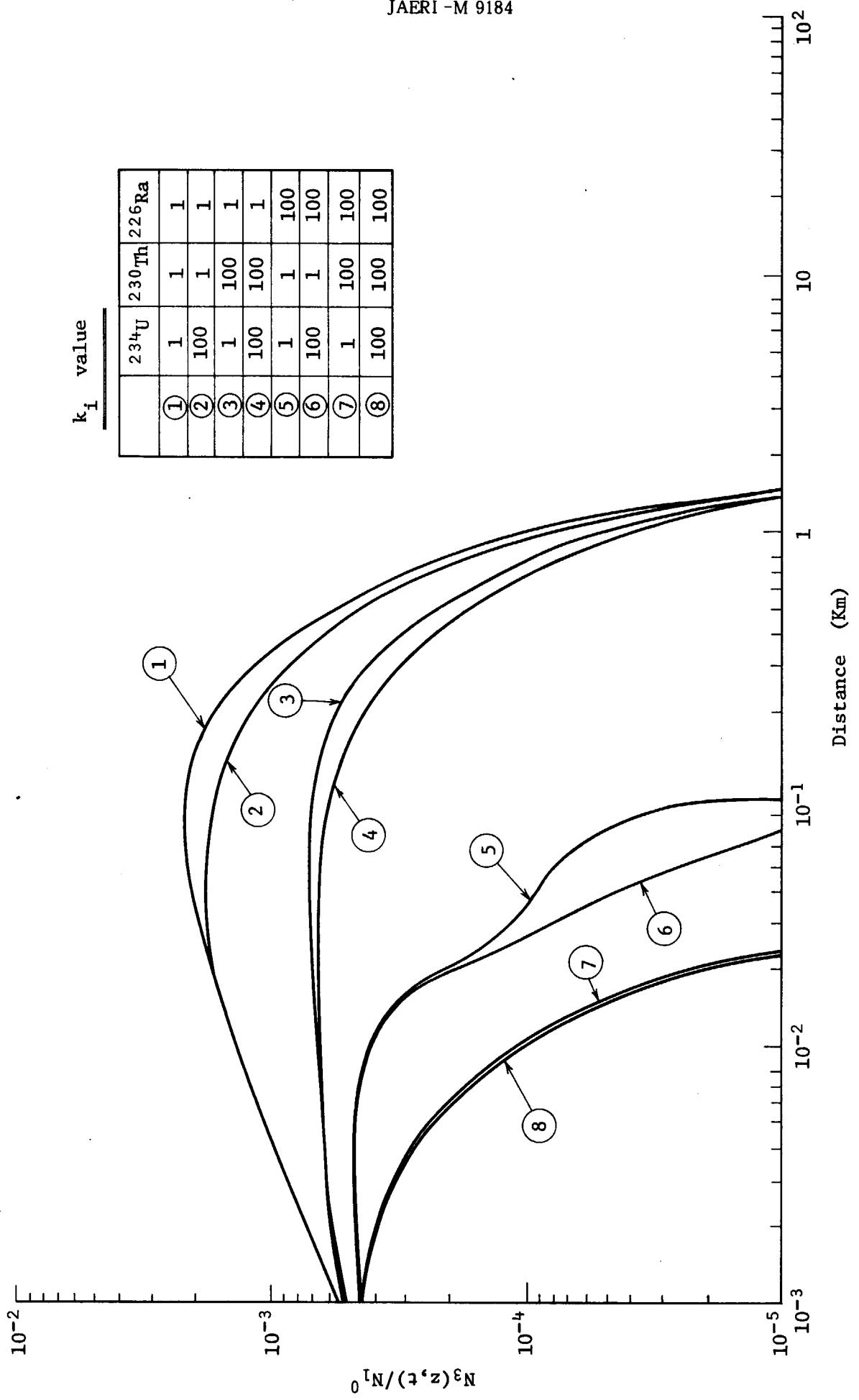


Fig. 3 Effect of mineralization of precursors on  $^{226}\text{Ra}$  concentration for the  $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$  decay chain at  $1 \times 10^4$  yr  
(Initial No Daughter)

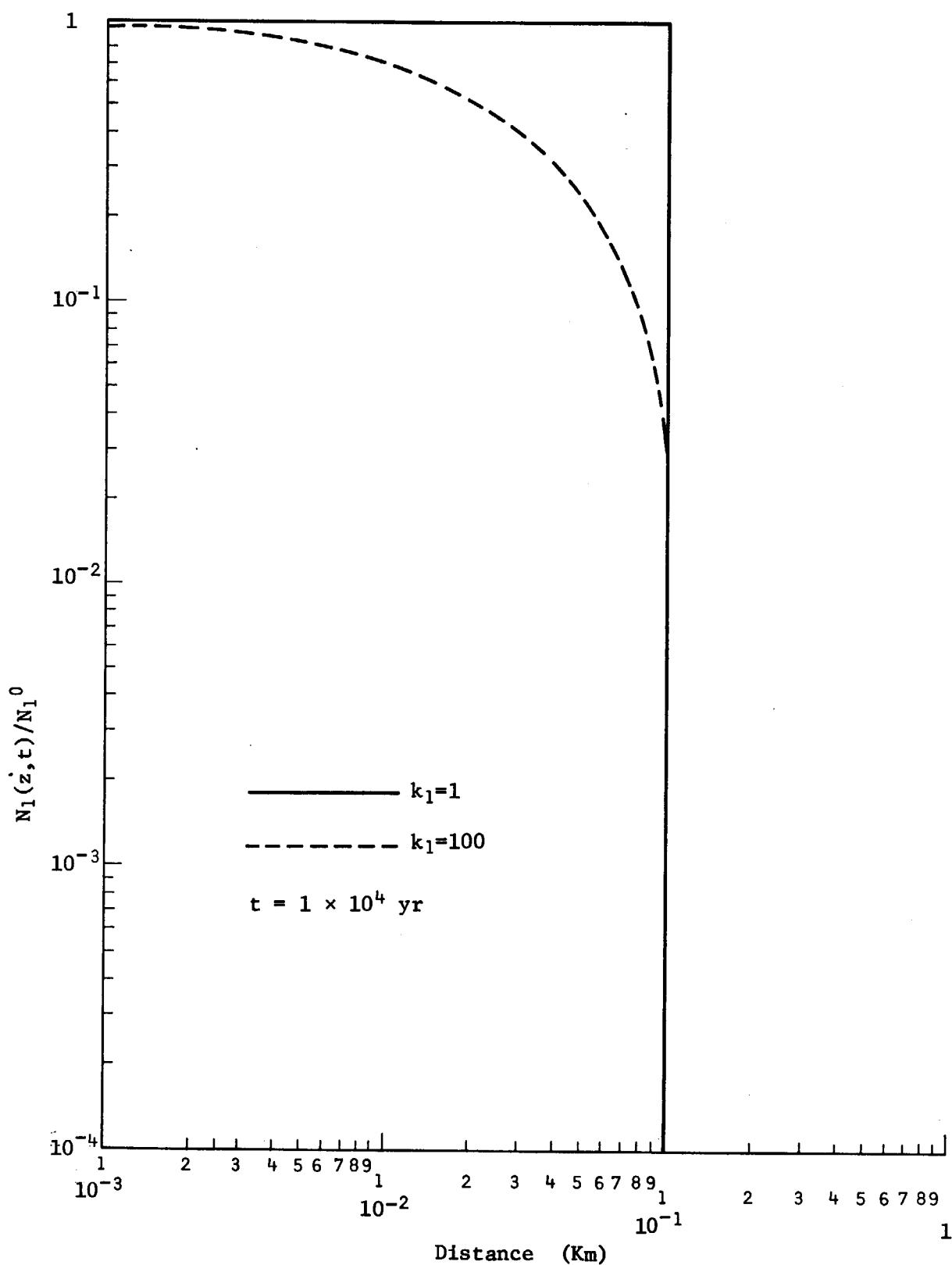
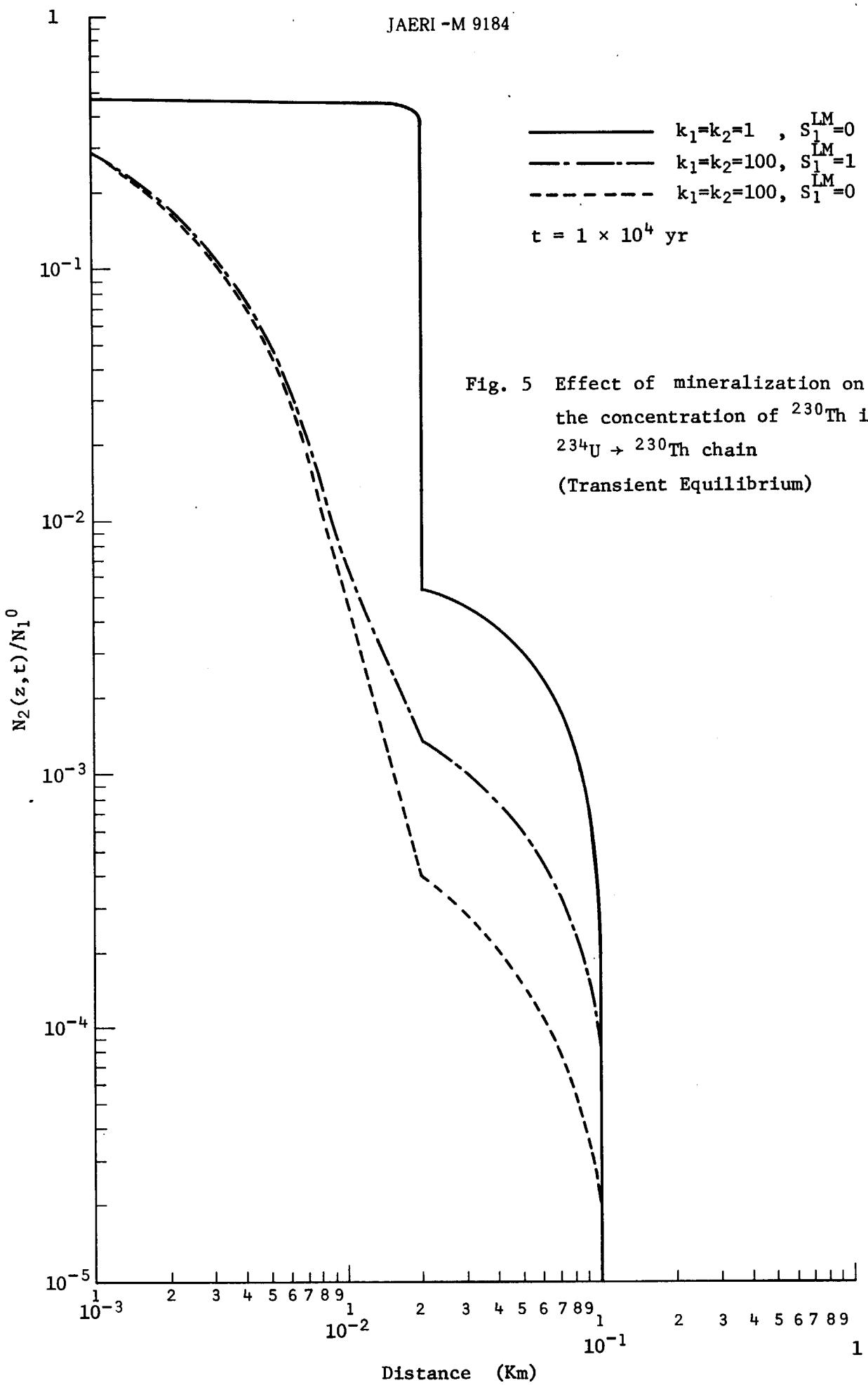
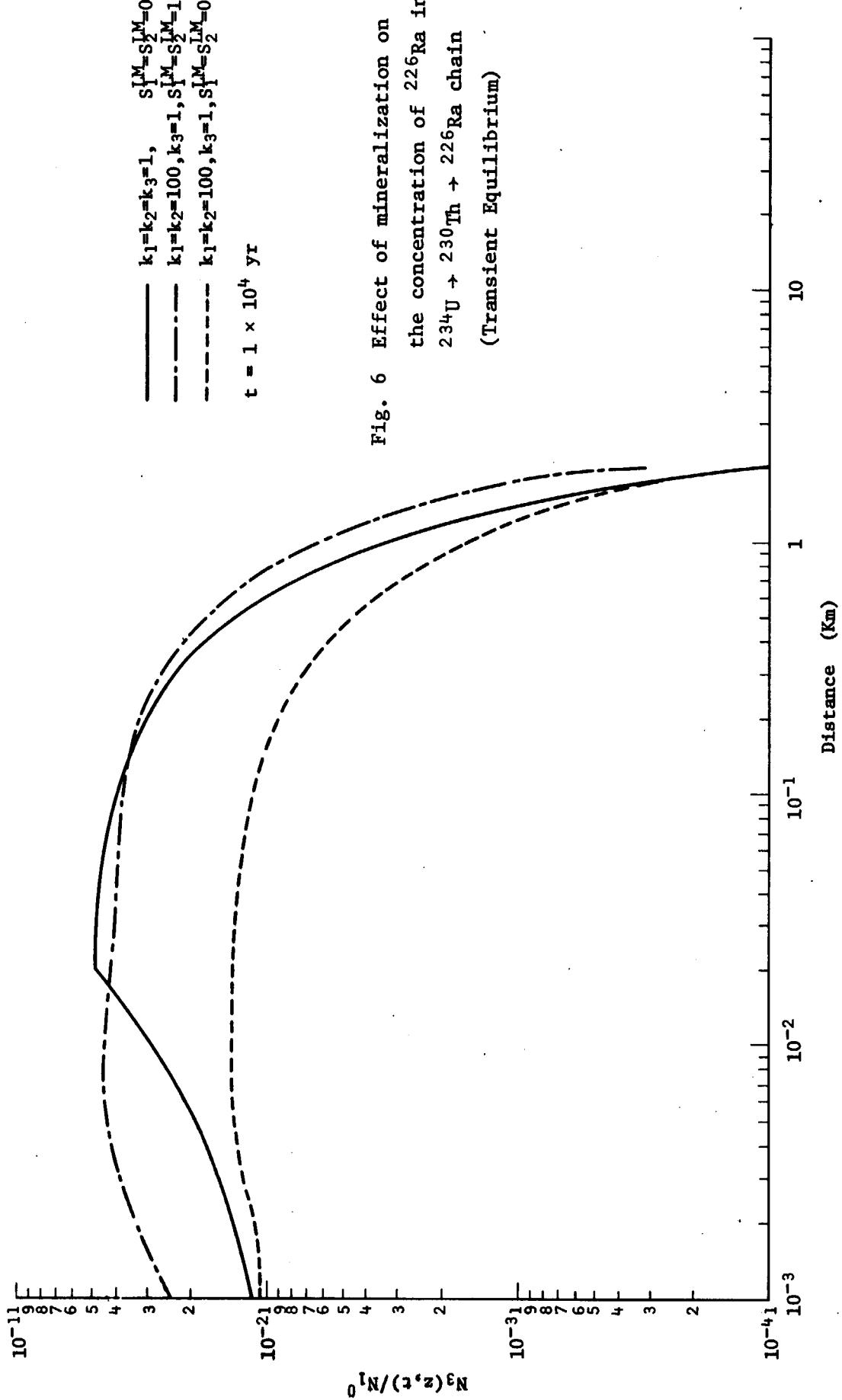


Fig. 4 Effect of mineralization on the concentration of  $^{234}\text{U}$   
(Transient Equilibrium)





## Appendix [1]

$$\text{where } \int_0^t e^{-\lambda_i(t-\theta)} L^{-1} \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_i}{\delta_i} d\theta$$

$$= \int_0^t e^{-\lambda_i(t-\theta)} e^{-\frac{z}{2\kappa}} L^{-1} \frac{-\frac{|z|}{2\kappa} \delta_i}{\delta_i} d\theta$$

$$\text{here } L^{-1} \frac{-\frac{|z|}{2\kappa} \delta_i}{\delta_i} = L^{-1} \frac{e^{-\frac{|z|}{2\kappa} \sqrt{1 + \frac{4\kappa(s+\lambda_i^*)}{v_i}}}}{\sqrt{1 + \frac{4\kappa(s+\lambda_i^*)}{v_i}}}$$

$$= L^{-1} \frac{1}{\sqrt{\frac{4\kappa}{v_i} \sqrt{s + (\lambda_i^* + \frac{v_i}{4\kappa})}}} e^{-\frac{|z|}{2\kappa} \sqrt{\frac{4\kappa}{v_i}} \sqrt{s + (\lambda_i^* + \frac{v_i}{4\kappa})}}$$

$$= \frac{1}{\sqrt{\frac{4\kappa}{v_i} \sqrt{\pi\theta}}} e^{-\frac{z^2}{4\kappa v_i \theta}} e^{-(\lambda_i^* + \frac{v_i}{4\kappa})\theta}$$

$$\therefore \int_0^t e^{-\lambda_i(t-\theta)} L^{-1} \frac{\frac{z}{2\kappa} - \frac{|z|}{2\kappa} \delta_i}{\delta_i} d\theta$$

$$= e^{\frac{z}{2\kappa}} e^{-\lambda_i t} \int_0^t e^{-\lambda_i \theta} \frac{1}{\sqrt{4\pi\kappa v_i \theta}} v_i e^{-\frac{z^2}{4\kappa v_i \theta}} e^{-(\lambda_i^* + \frac{v_i}{4\kappa})\theta} d\theta$$

$$= e^{\frac{z}{2\kappa}} e^{-\lambda_i t} \int_0^t e^{-(\lambda_i^* - \lambda_i + \frac{v_i}{4\kappa})\theta} \frac{e^{-\frac{z^2}{4\kappa v_i \theta}}}{\sqrt{4\pi\kappa v_i \theta}} v_i d\theta$$

$$\text{here let } \omega = \frac{v_i \theta}{4\pi}$$

$$= e^{\frac{z}{2\kappa}} e^{-\lambda_i t} \int_0^t e^{-\{1 + \frac{4\kappa(\lambda_i^* - \lambda_i)}{v_i}\}\omega} \frac{e^{-(\frac{z}{2\kappa})^2 \frac{1}{\omega}}}{\sqrt{\pi\omega}} d\omega$$

$$= e^{\frac{z}{2\kappa}} e^{-\lambda_i t} \frac{1}{2\sqrt{1 + \frac{4\kappa(\lambda_i^* - \lambda_i)}{v_i}}} [e^{-\frac{|z|}{2K} \sqrt{1 + \frac{4\kappa(\lambda_i^* - \lambda_i)}{v_i}}} \operatorname{erfc} \frac{|z| - v_i t \sqrt{1 + \frac{4\kappa(\lambda_i^* - \lambda_i)}{v_i}}}{\sqrt{4\kappa v_i t}}]$$

$$- e^{+\frac{z}{2K} \sqrt{1 + \frac{4\kappa(\lambda_i^* - \lambda_i)}{v_i}}} \operatorname{erfc} \frac{|z| + v_i t \sqrt{1 + \frac{4\kappa(\lambda_i^* - \lambda_i)}{v_i}}}{\sqrt{4\kappa v_i t}}$$

$$= E(i, i; i)$$

## Appendix [2]

$$\begin{aligned}
 I &= L_{\theta_1} L_{\theta_2} [e^{-\lambda_1^* \theta_1 - \lambda_2^* \theta_2} F(v_1 \theta_1 + v_2 \theta_2, z - v_1 \theta_1 - v_2 \theta_2)] \\
 &= \int_0^\infty \int_0^\infty e^{-(\lambda_1^* + s) \theta_1} e^{-(\lambda_2^* + s) \theta_2} F(v_1 \theta_1 + v_2 \theta_2, z - v_1 \theta_1 - v_2 \theta_2) d\theta_1 d\theta_2 \\
 &= \int_0^\infty \int_0^\infty e^{-(\lambda_1^* + s) \theta_1} e^{-(\lambda_2^* + s) \theta_2} \frac{e^{-(z - v_1 \theta_1 - v_2 \theta_2)^2 / 4\kappa(v_1 \theta_1 + v_2 \theta_2)}}{\sqrt{4\pi\kappa(v_1 \theta_1 + v_2 \theta_2)}} d\theta_1 d\theta_2
 \end{aligned}$$

here let

$$\left[ \begin{array}{l} x = v_1 \theta_1 + v_2 \theta_2 \\ y = v_1 \theta_1 \end{array} \right] \quad \left[ \begin{array}{l} \infty > x > y \\ \infty > y > 0 \end{array} \right]$$

$$\left[ \begin{array}{l} \therefore \theta_1 = \frac{y}{v_1}, \quad \theta_2 = \frac{1}{v_2} (x-y) \end{array} \right]$$

$$= \frac{1}{v_1 v_2} \int_0^\infty dy \int_y^\infty dx e^{-(\frac{\lambda_1^* + s}{v_1} - \frac{\lambda_2^* + s}{v_2})y} e^{-\frac{\lambda_2^* + s}{v_2} x} \frac{e^{-(z-x)^2 / 4\kappa x}}{\sqrt{4\pi\kappa x}}$$

$$\left[ \begin{array}{l} g_{12} = \frac{\lambda_1^* + s}{v_1} - \frac{\lambda_2^* + s}{v_2} \\ h_2 = \frac{\lambda_2^* + s}{v_2} \end{array} \right]$$

$$= \frac{1}{v_1 v_2} \int_0^\infty dy \int_y^\infty dx e^{-g_{12}y} e^{-h_2 x} \frac{e^{-(z-x)^2 / 4\kappa x}}{\sqrt{4\pi\kappa x}}$$

here let

$$\begin{aligned}
 I_1 &= \int_y^\infty e^{-h_2 x} \frac{e^{-(z-x)^2 / 4\kappa x}}{\sqrt{4\pi\kappa x}} dx \\
 &= \int_y^\infty \frac{1}{\sqrt{4\pi\kappa x}} e^{-\frac{z^2 + (1+4\kappa h_2)x^2}{4\kappa x}} + \frac{z}{2\kappa} dx
 \end{aligned}$$

$$[\delta_2 = \sqrt{1+4\kappa h_2}]$$

$$= \int_y^\infty \frac{1}{\sqrt{4\pi\kappa x}} e^{-\frac{z^2 + \delta_2^2 x^2}{4\kappa x}} + \frac{z}{4\kappa} dx$$

$$\left[ \begin{array}{l} \xi^2 = \frac{\delta_2^2 x^2}{4\kappa x} = \frac{\delta_2^2}{4\kappa} x \\ dx = 2\sqrt{x} d\xi / \sqrt{\frac{\delta_2^2}{4\kappa}} \end{array} \right]$$

(to be continued)

$$\begin{aligned}
&= \frac{1}{\sqrt{\pi}} e^{\frac{z}{2K}} \int_{\sqrt{\frac{\delta_2^2}{4K} y}}^{\infty} \frac{2}{\delta_2} e^{-(\xi^2 + \frac{z^2 \delta_2^2}{16K^2 \xi^2})} d\xi \\
&= \frac{e^{\frac{z}{2K}}}{\delta_2} \frac{1}{2} [2e^{-\frac{\delta_2}{2K}|z|} - e^{-\frac{\delta_2}{2K}|z|} \operatorname{erfc} \frac{|z|-y\delta_2}{\sqrt{4Ky}} + e^{\frac{\delta_2}{2K}|z|} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4Ky}}] \\
\therefore I &= \frac{1}{v_1 v_2} \left[ \int_0^{\infty} \frac{e^{\frac{z}{2K}} - \frac{|z|}{2K} \delta_2}{\delta_2} e^{-g_{12}y} dy - \int_0^{\infty} \frac{e^{\frac{z}{2K}} - \frac{|z|}{2K} \delta_2}{2\delta_2} \{e^{-\frac{|z|}{2K} \delta_2} e^{-g_{12}y} \operatorname{erfc} \frac{|z|-y\delta_2}{\sqrt{4Ky}} \right. \\
&\quad \left. - e^{-\frac{|z|}{2K} \delta_2} e^{-g_{12}y} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4Ky}}\} dy \right] \\
&= \frac{1}{v_1 v_2} \left[ \frac{e^{\frac{z}{2K}} - \frac{|z|}{2K} \delta_2}{\delta_2} \frac{e^{-g_{12}y}}{-g_{12}} \Big|_0^{\infty} - \int_0^{\infty} \frac{e^{\frac{z}{2K}} - \frac{|z|}{2K} \delta_2}{2\delta_2} \{e^{-\frac{|z|}{2K} \delta_2} e^{-g_{12}y} \operatorname{erfc} \frac{|z|-y\delta_2}{\sqrt{4Ky}} \right. \\
&\quad \left. - e^{-\frac{|z|}{2K} \delta_2} e^{-g_{12}y} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4Ky}}\} dy \right]
\end{aligned}$$

here let

$$\begin{aligned}
I_2 &= \int_0^{\infty} e^{-g_{12}y} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4Ky}} dy \quad - \frac{(|z|+y\delta_2)^2}{4Ky} \\
I_2 &= - \frac{e^{-g_{12}y}}{g_{12}} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4Ky}} \Big|_0^{\infty} + \frac{1}{g_{12}} \int_0^{\infty} e^{-g_{12}y} \left( -\frac{2}{\sqrt{\pi}} \frac{e^{-\frac{(|z|+y\delta_2)^2}{4Ky}}}{\sqrt{4Ky}} \right. \\
&\quad \times \left. \left( -\frac{|z|}{2y} \pm \frac{\delta_2}{2} \right) dy \right) \\
&[y\delta_2 = y', \quad \frac{K}{\delta_2} = K'] \\
&= - \frac{e^{-g_{12}y}}{g_{12}} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4Ky}} \Big|_0^{\infty} + \frac{1}{g_{12}} \int_0^{\infty} e^{-g_{12}y} \frac{y'}{\delta_2} \frac{1}{\sqrt{\pi}} \frac{e^{-\frac{(|z|+y')^2}{4K'y'}}}{\sqrt{4K'y'}} \left( \frac{|z|}{y'} \pm 1 \right) dy'
\end{aligned}$$

here let

$$I_3 = \frac{1}{\sqrt{\pi}} \int_0^{\infty} e^{-\left\{ \frac{g_{12}}{\delta_2} y' + \frac{(|z|+y')^2}{4K'y'} \right\}} \left( \frac{|z|}{\sqrt{4K'y'^3}} \pm \frac{1}{\sqrt{4K'y'}} \right) dy'$$

here

(to be continued)

$$\begin{aligned}
 & \frac{1}{\sqrt{\pi}} \int_0^\infty e^{-\left\{\frac{g_{12}}{\delta_2} y' + \frac{(|z|+y')^2}{\sqrt{4\kappa'y'}}\right\}} \frac{|z|}{\sqrt{4\kappa'y'^3}} dy' \\
 &= \frac{1}{\sqrt{\pi}} \int_0^\infty \frac{|z|}{\sqrt{4\kappa'y'^3}} e^{-\frac{z^2+2|z|y'+y'^2}{4\kappa'y'}} - \frac{g_{12}y'}{\delta_2} dy' \\
 &\quad \left[ \begin{array}{l} \eta = \frac{|z|}{\sqrt{4\kappa'y'}} \\ -2d\eta = \frac{|z|}{\sqrt{4\kappa'y'^3}} dy' \end{array} \right] \\
 &= -\frac{2}{\sqrt{\pi}} e^{\pm \frac{|z|}{2\kappa'}} \int_0^\infty e^{-[\eta^2 + (\frac{|z|}{4\kappa y})^2(1 + \frac{4\kappa' g_{12}}{\delta_2})]} d\eta \\
 &= -e^{\pm \frac{z}{2\kappa'}} - \frac{|z|}{2\kappa'} \sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}}
 \end{aligned}$$

$$\begin{aligned}
 & \frac{1}{\sqrt{\pi}} \int_0^\infty e^{-\left\{\frac{g_{12}}{\delta_2} y' + \frac{(|z|+y')^2}{\sqrt{4\kappa'y'}}\right\}} \frac{1}{\sqrt{4\kappa'y'}} dy' \\
 &= \frac{1}{\sqrt{\pi}} \int_0^\infty \frac{1}{\sqrt{4\kappa'y'}} dy' \\
 &\quad \left[ \begin{array}{l} \eta^2 = y' \frac{1 + \frac{4\kappa' g_{12}}{\delta_2}}{4\kappa'} \\ d\eta = \frac{1}{2\sqrt{4\kappa'y'}} \sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}} dy' \end{array} \right] \\
 &= \frac{e^{\pm \frac{z}{2\kappa'}}}{\sqrt{\pi}} \int_0^\infty \frac{2}{\sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}}} e^{-\{\eta^2 + (\frac{|z|}{4\kappa'})^2(1 + \frac{4\kappa' g_{12}}{\delta_2})\frac{1}{\eta^2}\}} d\eta \\
 &= e^{\pm \frac{|z|}{2\kappa'}} - \frac{|z|}{2\kappa'} \sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}} \Big/ \sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}} \\
 &\therefore I_3 = -e^{-\frac{|z|}{2\kappa'}(1-\alpha)} \pm \frac{e^{\frac{|z|}{2\kappa'}(1-\alpha)}}{\sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}}}
 \end{aligned}$$

$$\text{where } \alpha = \sqrt{1 + \frac{4\kappa' g_{12}}{\delta_2}}$$

(to be continued)

$$\begin{aligned}
\therefore I_2 &= -\frac{e^{-g_{12}y}}{g_{12}} \operatorname{erfc} \frac{|z|+y\delta_2}{\sqrt{4\kappa y}} \Big|_0^\infty \\
&\quad + \frac{e^{\frac{|z|}{2\kappa'}}}{2g_{12}} [2e^{-\frac{|z|}{2\kappa'}\alpha} - e^{\frac{|z|}{2\kappa'}\alpha} \operatorname{erfc} \frac{|z|+0}{\sqrt{0}} - e^{-\frac{|z|}{2\kappa'}\alpha} \operatorname{erfc} \frac{|z|-0}{\sqrt{0}}] \\
&\quad + \frac{e^{\pm\frac{|z|}{2\kappa'}}}{2g_{12}\alpha} [e^{-\frac{|z|}{2\kappa'}\alpha} - e^{-\frac{|z|}{2\kappa'}\alpha} \operatorname{erfc} \frac{|z|-0}{\sqrt{0}} + e^{\frac{|z|}{2\kappa'}\alpha} \operatorname{erfc} \frac{|z|+0}{\sqrt{0}}] \\
\therefore I &= \frac{e^{\frac{|z|}{2\kappa}}}{v_1 v_2} \left[ \frac{e^{-\frac{|z|}{2\kappa}\delta_2}}{\delta_2 g_{12}} e^{-g_{12}0} - \frac{e^{-\frac{|z|}{2\kappa}\delta_2}}{\delta_2 \cdot g\kappa} e^{-g_{12}\cdot\infty} \right. \\
&\quad \left. - \frac{e^{\frac{|z|}{2\kappa}}}{v_1 v_2} \frac{e^{-\frac{|z|}{2\kappa}\delta_2}}{2\delta_2} \left[ -\frac{2e^{-g_{12}\cdot\infty}}{g_{12}} + \frac{e^{-g_{12}\cdot 0}}{g_{12}} \operatorname{erfc} \frac{|z|-0}{\sqrt{0}} \right. \right. \\
&\quad \left. \left. + \frac{e^{(1-\alpha\delta_2)\frac{|z|}{2\kappa'}}}{g_{12}} + \frac{e^{(1-\alpha\delta_2)\frac{|z|}{2\kappa'}}}{g_{12}\alpha} \right] \right. \\
&\quad \left. + \frac{e^{\frac{|z|}{2\kappa}}}{v_1 v_2} \frac{e^{\frac{|z|}{2\kappa}}}{2\delta_2} \left[ \frac{e^{-g_{12}\cdot 0}}{g_{12}} \operatorname{erfc} \frac{|z|+0}{\sqrt{0}} + \frac{e^{-(1+\alpha\delta_2)\frac{|z|}{2\kappa'}}}{g_{12}} + \frac{e^{-(1+\alpha\delta_2)\frac{|z|}{2\kappa'}}}{g_{12}\alpha} \right] \right. \\
&\quad \left. = \frac{e^{\frac{|z|}{2\kappa}}}{v_1 v_2} \left[ \frac{e^{-\frac{|z|}{2\kappa}\delta_2}}{\delta_2 \cdot g_{12}} e^{-g_{12}\cdot 0} - \frac{-\frac{|z|}{2\kappa}\delta_2}{2\delta_2 g_{12} \cdot \alpha} e^{-\frac{|z|}{2\kappa'}\alpha} - \frac{|z|}{2\kappa'}\alpha \cdot \delta_2 \right. \right. \\
&\quad \left. \left. - \frac{e^{-\frac{|z|}{2\kappa}\delta_2}}{2\delta_2} \frac{e^{-\frac{|z|}{2\kappa'}\alpha}}{g_{12}\alpha} e^{-\frac{|z|}{2\kappa}\alpha\delta_2} \right] \right. \\
&\quad \left. = \frac{e^{\frac{|z|}{2\kappa}}}{v_1 v_2} \left[ \frac{e^{-\frac{|z|}{2\kappa}\delta_2}}{\delta_2 g_{12}} - \frac{e^{-\frac{|z|}{2\kappa}\alpha\delta_2}}{\delta_2 g_{12}\alpha} \right] \right]
\end{aligned}$$

here  $\delta_2 = \sqrt{1 + \frac{4\kappa(\lambda_2^*+s)}{v_2}}$ ,  $\alpha = \sqrt{1 + \frac{4\kappa'g_{12}}{\delta_2}} = \sqrt{1 + \frac{4\kappa g_{12}}{\delta_2^2}}$

$$\delta_2 \cdot \alpha = \sqrt{\alpha^2 + 4\kappa'g_{12}}$$

$$\begin{aligned}
&= \sqrt{1 + \frac{4\kappa(\lambda_2^*+s)}{v_2} + 4\kappa \left( \frac{\lambda_1^*+s}{v_1} - \frac{\lambda_2^*+s}{v_2} \right)} \\
&= \sqrt{1 + \frac{4\kappa(\lambda_1^*+s)}{v_1}}
\end{aligned}$$

(to be continued)

$$= \delta_1$$

$$I = \frac{\frac{1}{v_1 v_2} e^{\frac{z}{2\kappa}}}{\frac{\lambda_1^* + s}{v_1} - \frac{\lambda_2^* + s}{v_2}} \left[ \frac{e^{-\frac{|z|}{2\kappa}} \delta_2}{\delta_2} - \frac{e^{-\frac{|z|}{2\kappa}} \delta_1}{\delta_1} \right]$$

## Appendix [ 3 ] PROGRAM MINEL01

```

DIMENSION ANAME(5),HLIFE(5),CURIN(5),COEFK(5),T(2),Z(2),R(5),AT(5)
1      ,V(5),C2(5),C3(60),D1(2),D2(5,2),D3(15,2),CN(5),CM(5)
2      ,RCGW(3),FR(5),FK(5)
READ(5,501) IC,ICAL,IRCG,IDM
READ(5,501) (ANAME(I),HLIFE(I),CURIN(I),COEFK(I),RCGW(I),I=1,IC)
READ(5,502) DIF,VEL,FLW,TL
READ(5,503) IT,T(1),T(2)
READ(5,503) IZ,Z(1),Z(2)
READ(5,504) (FK(I),I=1,IC)
READ(5,505) SLM,SLL,SLN
500 FORMAT(4I1)
501 FORMAT(A7,4F8.0)
502 FORMAT(4F8.0)
503 FORMAT(I2,2F8.0)
504 FORMAT(3F10.0)
505 FORMAT(3F8.0)
DO 5 I=1,IC
A= ALOG(2.)/HLIFE(I)
R(I)= A
AT(I)= CURIN(I)/(FLW*TL)
IF(ICM.EQ.1) AT(I)= AT(I)/A
5 V(I)= VEL/CCEFK(I)
VMAX= AMAX1(V(1),V(2),V(3))
VMIN= AMIN1(V(1),V(2),V(3))
CUR= R(1)*CURIN(1)/1.16683E18
IF(ID1.EQ.1) CUR= CURIN(1)
D= DIF/VEL
WRITE(6,605) (ANAME(I),HLIFE(I),CURIN(I),COEFK(I),R(I),V(I),AT(I),
1I=1,IC)
WRITE(6,606) DIF,VEL,FLW,TL
WRITE(6,607) (FK(I),I=1,IC)
WRITE(6,608) SLM,SLL,SLN
605 FORMAT(10X,A7,1P4E15.3,*YR*,5E15.3)
606 FORMAT(10X,1P4E15.3,1H1)
607 FORMAT(3F10.0)
608 FORMAT(3F8.0)
R12= R(1)-R(2)
R21= R(2)-R(1)
R23= R(2)-R(3)
R31= R(3)-R(1)
RR= R(1)*R(2)*AT(1)
B11= AT(1)
B12= -AT(1)*R(1)/R12
B22= AT(2)-B12
B13= -RR/(R12*R31)
B23= -RF/(R12*R23)-AT(2)*R(2)/R23
B33= -RR/(R23*R31)+AT(2)*R(2)/R23+AT(3)
D4= 4.*D
S1= 1./SQRT(V(1)*D)
S2= 1./SQRT(V(2)*D)
S3= 1./SQRT(V(3)*D)
DO 6 I=1,IC
6 FR(I)=R(I)*FK(I)
F12=(V(1)*FR(2)-V(2)*FR(1))/(V(1)-V(2))
F23=(V(2)*FR(3)-V(3)*FR(2))/(V(2)-V(3))
F31=(V(3)*FR(1)-V(1)*FR(3))/(V(3)-V(1))

```

```

V1= V(1)/D4
V2= V(2)/D4
V3= V(3)/D4
IX= 0
TD= 0.
10 CCNTINUE
E1= EXP(-R(1)*TC)
E2= EXP(-R(2)*TD)
E3= EXP(-R(3)*TC)
B11= B11*E1
B12= B12*E1
B13= B13*E1
B22= B22*E2
B23= B23*E2
B33= B33*E3
RAM12= FF(1)/V(1)-FR(2)/V(2)
RAM23= FR(2)/V(2)-FR(3)/V(3)
RAM31= FR(3)/V(3)-FR(1)/V(1)
RAM21= -RAM12
RAM32= -RAM23
RAM13= -PAM31
GAM12= 1.0/V(1)-1.0/V(2)
GAM23= 1.0/V(2)-1.0/V(3)
GAM31= 1.0/V(3)-1.0/V(1)
GAM21= -GAM12
GAM32= -GAM23
GAM13= -GAM31
BT12= PAM12/CAM12
BT23= RAM23/GAM23
BT31= RAM31/GAM31
BT21= BT12
BT32= BT23
BT13= BT31
C1= B11
C2(1)= B12
C2(2)= B22
C2(3)= -B11*R(1)/(V(1)*RAM12-V(1)*R(1)*GAM12)
C2(4)= B11*SLN*R(1)*(FR(1)-R(1))*V(2)/((BT12-R(1))**2)*
1 (V(1)-V(2))
C3(1)= B13
C3(2)= B23
C3(3)= B33
C3(4)= B12*R(2)/(V(2)*RAM23-V(2)*R(1)*GAM23)
C3(5)= -B22*R(2)/(V(2)*RAM23-V(2)*R(2)*GAM23)
C3(6)= B11*R(1)*R(2)/((V(1)*V(2))*(RAM32-R(1)*GAM32)*(RAM12-R(1)*
1 GAM12))
C3(7)= B11*R(1)*R(2)/((V(1)*V(2))*(RAM13-R(1)*GAM13)*
1 (RAM22-R(1)*GAM23))
C3(8)= B11*R(1)*R(2)/((V(1)*V(2))*(RAM31-R(1)*GAM31)*
1 (RAM21-R(1)*GAM21))
C3(9)= -B11*R(1)*R(2)*GAM32/((V(1)*V(2))*(RAM32-R(1)*GAM32)*
1 (PAM12*GAM32-RAM32*GAM12))
C3(10)= E11*P(1)*R(2)*GAM12/((V(1)*V(2))*(RAM12-R(1)*GAM12)*
1 (PAM12*GAM32-RAM32*GAM12))
C3(11)= -B11*R(1)*P(2)*GAM31/((V(1)*V(2))*(RAM31-R(1)*GAM31)*
1 (PAM21*GAM31-RAM31*GAM21))
C3(12)= B11*P(1)*R(2)*GAM21/((V(1)*V(2))*(RAM21-R(1)*GAM21)*
1 (PAM21*GAM31-RAM31*GAM21))

```

$C3(13) = -B11*R(1)*R(2)*GAM13/((V(1)*V(2))*(RAM13-R(1)*GAM13)*$   
 1  $(RAM23*GAM13-RAM13*GAM23))$   
 $C3(14) = B11*R(1)*R(2)*GAM23/((V(1)*V(2))*(RAM23-R(1)*GAM23)*$   
 1  $(RAM23*GAM13-RAM13*GAM23))$   
 $C3(16) = (B11*SLM*R(1)*R(2)*(FR(1)-R(2))/V(1)*V(2))/$   
 1  $GAM13*GAM23*((R(1)-BT13)**2)*(BT23-BT13)$   
 $C3(17) = (B11*SLM*R(1)*R(2)*(FR(1)-R(2))/V(1)*V(2))/$   
 1  $GAM13*GAM23*((R(1)-BT23)**2)*(BT13-BT23)$   
 $C3(19) = (B11*SLM*R(1)*R(2)*(FR(1)-R(2))/V(1)*V(2))/$   
 1  $GAM32*GAM12*((R(1)-BT32)**2)*(BT12-BT32)$   
 $C3(20) = (B11*SLM*R(1)*R(2)*(FR(1)-R(2))/V(1)*V(2))/$   
 1  $GAM32*GAM12*((R(1)-BT12)**2)*(BT32-BT12)$   
 $C3(22) = (B11*SLM*R(1)*R(2)*(FR(1)-R(2))/V(1)*V(2))/$   
 1  $GAM21*GAM31*((R(1)-BT21)**2)*(BT31-BT21)$   
 $C3(23) = (B11*SLM*R(1)*R(2)*(FR(1)-R(2))/V(1)*V(2))/$   
 1  $GAM21*GAM31*((R(1)-BT31)**2)*(BT21-BT31)$   
 $C3(25) = B11*SLM*SLL*R(1)*R(2)*(FR(1)-R(1))*V(3)/(V(1)-V(3))*$   
 1  $((R12**2)*(BT31-R(2)))$   
 $C3(26) = B11*SLM*SLL*R(1)*R(2)*(FR(1)-R(1))*V(3)/(V(1)-V(3))*$   
 1  $((R(1)-BT31)**2)*(R(2)-BT31)$   
 $C3(27) = B22*SLM*R(2)*(FR(2)-R(2))*V(3)/(V(2)-V(3))*$   
 1  $((BT23-R(2))**2)$   
 $C3(29) = -B12*SLM*R(2)*(FR(2)-R(2))*V(3)/(V(2)-V(3))*R12*$   
 1  $((R(2)-BT23))$   
 $C3(30) = -B12*SLM*R(2)*(FR(2)-R(2))*V(3)/(V(2)-V(3))*$   
 1  $R12*(BT23-R(1))$   
 $C3(31) = -B12*SLM*R(2)*(FR(2)-R(2))*V(3)/(V(2)-V(3))*$   
 1  $((BT23-R(1))*(R(2)-BT23))$   
 $C3(32) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM13*GAM23*R21*(BT13-R(1))*(BT23-R(1))$   
 $C3(33) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM13*GAM23*R12*(BT13-R(2))*(BT23-R(2))$   
 $C3(34) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM13*GAM23*(R(1)-BT13)*(R(2)-BT13)*(BT23-BT13)$   
 $C3(35) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM13*GAM23*(R(1)-BT23)*(R(2)-BT23)*(BT13-BT23)$   
 $C3(36) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM32*GAM12*R21*(BT32-R(1))*(BT12-R(1))$   
 $C3(37) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM32*GAM12*R12*(BT32-R(2))*(BT12-R(2))$   
 $C3(38) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM32*GAM12*(R(1)-BT32)*(R(2)-BT32)*(BT12-BT32)$   
 $C3(39) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM32*GAM12*(R(1)-BT12)*(R(2)-BT12)*(BT32-BT12)$   
 $C3(40) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM21*GAM31*R21*(BT21-R(1))*(BT31-R(1))$   
 $C3(41) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM21*GAM31*R12*(BT21-R(2))*(BT31-R(2))$   
 $C3(42) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM21*GAM31*(R(1)-BT21)*(R(2)-BT21)*(BT31-BT21)$   
 $C3(43) = B11*SLN*R(1)*R(2)*(FR(2)-R(2))/V(1)*V(2)*$   
 1  $GAM21*GAM31*(R(1)-BT31)*(R(2)-BT31)*(BT21-BT31)$   
 $C3(45) = B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/$   
 1  $V(1)*V(2)*GAM13*GAM23*(R12**2)*(BT13-R(2))*(BT23-R(2))$   
 $C3(46) = B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/$   
 1  $V(1)*V(2)*GAM13*GAM23*(R(1)-BT13)**2)*(R(2)-BT13)*$   
 2  $(BT23-BT13)$   
 $C3(47) = B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/$

```

1      V(1)*V(2)*GAM13*GAM23*((R(1)-BT23)**2)*(R(2)-BT23)*
2      (BT13-BT23)
C3(49)= B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/_
1      V(1)*V(2)*GAM32*GAM12*(R12**2)*(BT32-R(2))*(BT12-R(2))
C3(50)= B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/_
1      V(1)*V(2)*GAM32*GAM12*((R(1)-BT32)**2)*(R(2)-BT32)*
2      (BT12-BT32)
C3(51)= B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/_
1      V(1)*V(2)*GAM32*GAM12*((R(1)-BT12)**2)*(R(2)-BT12)*
2      (BT32-BT12)
C3(53)= B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/_
1      V(1)*V(2)*GAM21*GAM31*(R12**2)*(BT21-R(2))*(BT31-R(2))
C3(54)= B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/_
1      V(1)*V(2)*GAM21*GAM31*((R(1)-BT21)**2)*(R(2)-BT21)*
2      (BT31-BT21)
C3(55)= B11*SLM*SLN*R(1)*R(2)*(FR(1)-R(1))*(FR(2)-R(2))/_
1      V(1)*V(2)*GAM21*GAM31*((R(1)-BT31)**2)*(R(2)-BT31)*
2      (BT21-BT31)
IF(IX.EQ.1) GO TO 13
D1(1)= C1
DO 11 I=1,3
11 D2(I,1)= C2(I)
DO 12 I=1,14
12 D3(I,1)= C3(I)
IX= 1
TD= TL
GO TO 10
13 D1(2)= C1
DO 14 I=1,3
14 D2(I,2)= C2(I)
DO 15 I=1,14
15 D3(I,2)= C3(I)
- WRITE(6,650) (C1(I),(D2(J,I),J=1,3),I=1,2)
WRITE(6,650) ((D3(J,I),J=1,14),I=1,2)
550 FORMAT(10E11.3)
IF(ICAL.EQ.1) GO TO 21
KM= 0
IZ1= IZ+1
IT1= IT+1
DO 100 M=1,IT1
T2= T(1)*10.**(M-1)
KM= 1
GO TO 20
16 CONTINUE
T2= T(1)*10.**(M-1)*5.
KM= 0
20 CONTINUE
T1= T2
WRITE(6,601) T2,(ANAME(I),I=1,3),(ANAME(I),I=1,3)
601 FORMAT(//,8X,*T=*,1PE10.3,15X,3(2X,A7,1X),5X,3(2X,A7,1X))
DO 90 N=1,IZ1
DO 90 L=1,N0
FL= L
Z1= Z(1)*10.**(N-1)*(1.+1*(FL-1.))
Z2= Z1/D4*2.
IA= 1
30 C1= D1(IA)
DO 31 I=1,3

```

---

31 C2(I) = D2(I, IA)  
DO 32 I=1, 14

---

32 C3(I) = D3(I, IA)  
IF(IA.EQ.2) T1= T2-TL

---

XA= ELF(R(1), FR(1)-R(1)+V1, Z1\*S1, Z2, T1)  
XB= ELF(R(1), FR(2)-R(1)+V2, Z1\*S2, Z2, T1)  
XC= ELF(R(2), FR(2)-R(2)+V2, Z1\*S2, Z2, T1)  
XD= ELF(F12, FR(1)-F12+V1, Z1\*S1, Z2, T1)

---

XE= ELF(F12, FR(2)-F12+V2, Z1\*S2, Z2, T1)  
XF= ELF(R(1), FR(3)-R(1)+V3, Z1\*S3, Z2, T1)  
XG= ELF(R(2), FR(3)-R(2)+V3, Z1\*S3, Z2, T1)  
XH= ELF(R(3), FR(3)-R(3)+V3, Z1\*S3, Z2, T1)

---

XI= ELF(F23, FR(2)-F23+V2, Z1\*S2, Z2, T1)  
XJ= ELF(F23, FR(3)-F23+V3, Z1\*S3, Z2, T1)

---

XK= ELF(F31, FR(1)-F31+V1, Z1\*S1, Z2, T1)  
XL= ELF(F31, FR(3)-F31+V3, Z1\*S3, Z2, T1)

---

XM= ELF(R(2), FR(1)-R(2)+V1, Z1\*S1, Z2, T1)

---

C2(5)= B11\*SLM\*R(1)\*(FR(1)-R(1))\*V(2)\*(1.-(R(1)-BT12)\*T1)/  
1 ((BT12-R(1))\*2)\*(V(1)-V(2))

---

C3(15)= (B11\*SLM\*R(1)\*R(1)\*(FR(1)-R(2))/V(1)\*V(2))\*  
1 ((T1/(BT13-R(1))\*(BT23-R(1))-((BT13-R(1))+(BT23-R(1))))/  
2 ((BT13-R(1))\*(BT23-R(1)))\*\*2)/GAM13\*GAM23)

---

C3(18)= (B11\*SLM\*R(1)\*R(1)\*(FR(1)-R(2))/V(1)\*V(2))\*  
1 ((T1/(BT32-R(1))\*(BT12-R(1))-((BT32-R(1))+(BT12-R(1))))/  
2 ((BT32-R(1))\*(BT12-R(1)))\*\*2)/GAM32\*GAM12)

---

C3(21)= (B11\*SLM\*R(1)\*R(1)\*(FR(1)-R(2))/V(1)\*V(2))\*  
1 ((T1/(BT21-R(1))\*(BT31-R(1))-((BT21-R(1))+(BT31-R(1))))/  
2 ((BT21-R(1))\*(BT31-R(1)))\*\*2)/GAM21\*GAM31)

---

C3(24)= B11\*SLM\*SLL\*R(1)\*R(2)\*(FR(1)-R(1))\*V(3)\*  
1 (T1/R21\*(BT31-R(1))-(R21-R(1)+BT31)/(R21\*(BT31-R(1)))\*\*2)  
2 /(V(1)-V(3))

---

C3(28)= -B22\*SLM\*R(2)\*(FR(2)-R(2))\*V(3)\*(1.-(R(2)-BT23)\*T1)/  
1 (V(2)-V(3))\*((BT23-R(2))\*2)

---

C3(44)= (B11\*SLM\*SLN\*R(1)\*R(2)\*(FR(1)-R(1))\*(FR(2)-R(2))/  
1 V(1)\*V(2)\*GAM13\*GAM23)\*(T1/R21\*(BT13-R(1))\*(BT23-R(1))/  
2 +(R21\*(BT13-R(1))+(BT13-R(1))\*(BT23-R(1))+(BT23-R(1))\*  
3 R21)/(R21\*(BT13-R(1))\*(BT23-R(1)))\*\*2)

---

C3(48)= (B11\*SLM\*SLN\*R(1)\*R(2)\*(FR(1)-R(1))\*(FR(2)-R(2))/  
1 V(1)\*V(2)\*GAM32\*GAM12)\*(T1/R21\*(BT32-R(1))\*(BT12-R(1))/  
2 +(R21\*(BT32-R(1))+(BT32-R(1))\*(BT12-R(1))+(BT12-R(1))\*  
3 P21)/(R21\*(BT32-R(1))\*(BT12-R(1)))\*\*2)

---

C3(52)= (B11\*SLM\*SLN\*R(1)\*R(2)\*(FR(1)-R(1))\*(FR(2)-R(2))/  
1 V(1)\*V(2)\*GAM21\*GAM31)\*(T1/R21\*(BT21-R(1))\*(BT31-R(1))/  
2 +(R21\*(BT21-R(1))+(BT21-R(1))\*(BT31-R(1))+(BT31-R(1))\*  
3 R21)/(R21\*(BT21-R(1))\*(BT31-R(1)))\*\*2)

---

CN(1)= .5\*C1\*XA  
CN(2)= .5\*(C2(1)\*XB+C2(2)\*XC+C2(3)\*(-XB+XA-XD+XE))  
1 +C2(4)\*(XA-XB)+C2(5)\*(-XD+XE))

---

YA= C3(1)\*XF+C3(2)\*XG+C3(3)\*XH+C3(4)\*(XF-XB+XI-XJ)  
1 +C3(5)\*(XC-XG+XJ-XI)+C3(6)\*XB+C3(7)\*XF+C3(8)\*XA  
2 +C3(9)\*XI+C3(10)\*XE+C3(11)\*XK+C3(12)\*XD  
3 +C3(13)\*XL+C3(14)\*XJ+C3(15)\*XF+C3(16)\*XL+C3(17)\*XJ  
4 +C3(18)\*XB+C3(19)\*XI+C3(20)\*XE+C3(21)\*XA+C3(22)\*XD

---

YB= C3(23)\*XK+C3(24)\*(XA-XF)+C3(25)\*(XM-XG)+C3(26)\*(XK-XL)  
1 +C3(27)\*(XC-XG)+C3(28)\*(XI-XJ)+C3(29)\*(XC-XG)  
2 +C3(30)\*(XR-XF)+C3(31)\*(XI-XJ)  
3 +C3(32)\*XF+C3(33)\*XG+C3(34)\*XL+C3(35)\*XJ+C3(36)\*XB  
4 +C3(37)\*XC+C3(38)\*XI+C3(39)\*XE+C3(40)\*XA+C3(41)\*XM

---

```

YC= C3(42)*XD+C3(43)*XK
1 +C3(44)*XF+C3(45)*XC+C3(46)*XL+C3(47)*XJ
2 +C3(48)*XB+C3(49)*XC+C3(50)*XI+C3(51)*XE
3 +C3(52)*XA+C3(53)*XM+C3(54)*XD+C3(55)*XK
CN(3)= .5*(YA+YB+YC)
IF(T2.LT.TL) GO TO 36
IF(IA.EQ.2) GO TO 34
DO 33 I=1,3
33 CM(I)= CN(I)
IA= 2
GO TO 30
34 DO 35 I=1,3
35 CN(I)= CM(I)-CN(I)
36 CONTINUE
DO 37 I=1,3
CM(I)= CN(I)/AT(I)
37 CN(I)= CN(I)*R(I)/(AT(I)*R(I))
CMAX= AMAX1(CN(1),CN(2),CN(3))
IF(IRCG.NE.1) GO TO 75
DC 38 I=1,3
CM(I)= CUR*CN(I)/TL
38 CN(I)= CM(I)/RCGW(I)
75 CONTINUE
ZMAX= VMAX*T2
IF(ZMAX.GT.Z1) GO TO 80
IF(CMAX.LT.1.E-100) GO TO 95
80 CONTINUE
WRITE(6,603) Z1,(CM(I),I=1,3),(CN(I),I=1,3)
603 FORMAT(20X,1PE10.3,5X,3E10.3,5X,3E10.3)
T1= T2
90 CONTINUE
95 CONTINUE
IF(KM.EQ.1) GO TO 13
100 CONTINUE
GO TO 300
21 KM= 0
IZ1= IZ+1
IT1= IT+1
DO 200 M=1,IZ1
JJ= 1
DO 250 J=1,4
JJ= JJ+J-1
FJ= JJ
Z1= Z(1)*10.**(M-1)*FJ
Z2= Z1/D4*2.C
WRITE(6,602) Z1,(ANAME(I),I=1,3),(ANAME(I),I=1,3)
602 FORMAT(//,8X,*Z=*,1PE10.3,15X,3(2X,A7,1X),5X,3(2X,A7,1X))
DO 190 N=1,IT1
DO 190 L=1,SC
FL= L
T2= T(1)*10.**(N-1)*(1.+1*(FL-1.))
DELT= ABS(T2-TL)
IF(DELT.LT.10.) GO TO 190
T1= T2
IA= 1
301 C1= D1(IA)
DO 311 I=1,3
311 C2(I)= D2(I,IA)

```

---

DC 321 I=1,14

---

321 C3(I)= D3(I,IA)

---

IF(IA.EQ.2) T1= T2-TL  
XA= ELF(R(1),FR(1)-P(1)+V1,Z1\*S1,Z2,T1)

---

IF(IC.EQ.1) GC TO 325  
XB= ELF(R(1),FR(2)-R(1)+V2,Z1\*S2,Z2,T1)

---

XC= ELF(R(2),FR(2)-R(2)+V2,Z1\*S2,Z2,T1)  
XD= ELF(F12,FR(1)-F12+V1,Z1\*S1,Z2,T1)

---

XE= ELF(F12,FR(2)-F12+V2,Z1\*S2,Z2,T1)  
XF= ELF(R(1),FR(3)-R(1)+V3,Z1\*S3,Z2,T1)

---

XG= ELF(R(2),FR(3)-R(2)+V3,Z1\*S3,Z2,T1)  
XH= ELF(R(3),FR(3)-R(3)+V3,Z1\*S3,Z2,T1)

---

XI= ELF(F23,FR(2)-F23+V2,Z1\*S2,Z2,T1)  
XJ= ELF(F23,FR(3)-F23+V3,Z1\*S3,Z2,T1)

---

XK= ELF(F31,FR(1)-F31+V1,Z1\*S1,Z2,T1)  
XL= ELF(F31,FR(3)-F31+V3,Z1\*S3,Z2,T1)

---

XM= ELF(R(2),FR(1)-R(2)+V1,Z1\*S1,Z2,T1)

---

325 CONTINUE

---

CN(1)= .5\*C1\*XA  
IF(IC.EQ.1) GC TO 326

---

CN(2)= .5\*(C2(1)\*XB+C2(2)\*XC+C2(3)\*(-XB+XA-XD+XE))  
1 +C2(4)\*(XA-XB)+C2(5)\*(-XD+XE))

---

YA= C3(1)\*XF+C3(2)\*XG+C3(3)\*XH+C3(4)\*(XF-XB+XI-XJ)  
1 +C3(5)\*(XC-XG+XJ-XI)+C3(6)\*XB+C3(7)\*XF+C3(8)\*XA

---

2 +C3(9)\*XI+C3(10)\*XE+C3(11)\*XK+C3(12)\*XD  
3 +C3(13)\*XL+C3(14)\*XJ+C3(15)\*XF+C3(16)\*XL+C3(17)\*XJ

---

4 +C3(18)\*XE+C3(19)\*XI+C3(20)\*XE+C3(21)\*XA+C3(22)\*XD  
YB= C3(23)\*XK+C3(24)\*(XA-XF)+C3(25)\*(XM-XG)+C3(26)\*(XK-XL)

---

1 +C3(27)\*(XC-XG)+C3(28)\*(XI-XJ)+C3(29)\*(XC-XG)  
2 +C3(30)\*(XR-XF)+C3(31)\*(XI-XJ)

---

3 +C3(32)\*XF+C3(33)\*XG+C3(34)\*XL+C3(35)\*XJ+C3(36)\*XB  
4 +C3(37)\*XC+C3(38)\*XI+C3(39)\*XE+C3(40)\*XA+C3(41)\*XM

---

YC= C3(42)\*XD+C3(43)\*XK  
1 +C3(44)\*XF+C3(45)\*XG+C3(46)\*XL+C3(47)\*XJ  
2 +C3(48)\*XE+C3(49)\*XC+C3(50)\*XI+C3(51)\*XE  
3 +C3(52)\*XA+C3(53)\*X4+C3(54)\*XD+C3(55)\*XK

---

CN(3)= .5\*(YA+YB+YC)

---

326 CONTINUE

---

IF(T2.LT.TL) GC TO 361  
IF(IA.EQ.2) GC TO 341

---

DO 331 I=1,IC

---

331 CM(I)= CN(I)

---

IA= 2  
GO TO 301

---

341 DO 351 I=1,IC

---

351 CN(I)= CM(I)-CN(I)

---

361 CONTINUE

---

DO 371 I=1,IC

---

CM(I)= CN(I)/AT(1)

---

371 CN(I)= CN(I)\*R(I)/(AT(1)\*R(I))

---

IF(IC.NE.1) GC TO 185  
CM(2)= 0.0

---

CM(3)= 0.0  
CN(2)= 0.0

---

CN(3)= 0.0

---

185 CONTINUE

---

CMAX= AMAX1(CN(1),CN(2),CN(3))

---

IF(IRCG.NE.1) GO TO 175

---

```
DO 381 I=1,IC
CM(I)= CUR*CN(I)/TL
381 CN(I)= CM(I)/RCGW(I)
175 CONTINUE
TMAX= Z1/VMIN+TL
IF(TMAX.GT.T2) GO TO 180
IF(CMAX.LT.1.E-100) GO TO 250
180 CONTINUE
WRITE(6,603) T2,(CM(I),I=1,3),(CN(I),I=1,3)
190 CONTINUE
250 CONTINUE
200 CONTINUE
300 CONTINUE
STOP
END
```

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