A Semi-analytical Solution for Advection-dispersion Migration of Radionuclides through Two-layered Geologic Media

By

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Abstract

A semi-analytical solution was obtained for a one-dimensional migration of radionuclides in four (or less) member-decay chains with dispersion through a two-layered sorbing medium. The Preferential-release model is applied, in which each element can take a different and time-depending release rate from the waste form. The two types of geologic media concerned were characterized by different retardation factors for each element, the dispersion coefficients, and the groundwater velocities between the two layers.

In connection with a long-term safety assessment of geologic disposal of high-level radioactive wastes (HLW), the concentration profiles of radionuclides predicted by the solution can be applied to heterogeneous geologic formations, in which radionuclides migrate through e. g. granite of a host rock followed by subsoil bounded by a surface water body.

I. Introduction

In a long-term safety assessment on the geologic disposal of high-level radioactive waste (HLW), mathematical models and corresponding computer codes describing the nuclide transport through geologic media are essential instruments. Radionuclide migration in complicated geologic systems has recently been able to be predicted by numerical models involving such situations as multi-dimensional transport, layered medium, and various rock-water interactions. Analytical models, on the other hand, play a role as a benchmark to check the accuracy of the numerical analyses.

Although some analytical or semi-analytical solutions for migration of radionuclides in decay chains have been presented for homogeneous geologic media[1-4], the solution for heterogeneous geologic media (layered media) has been obtained only by Hadermann and Patry[5], in which a simple assumption was placed on the release model of nuclides from the waste form.

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This paper presents the solution for migration of radionuclides in decay chains in the second layer followed by the first layer of host rock in which HLW will be buried. In the solution, the Preferential-release model[6] was applied to the release model of nuclides from the waste form. This release model may be much more general and a little more realistic than the Impulse-release model adopted by Hadermann and Patry[5].

II. Analysis

Although the migration behavior of radionuclides through geologic media depends on various phenomena, an extremely simplified model is used to obtain analytical solutions. Here, groundwater transport, dispersion, sorption, and decay chain are taken into account. Assuming a one-dimensional column for a multi-layered path of groundwater, the fundamental differential equation on the migration of radionuclide i in a decay chain of $A(i=1)\rightarrow B(i=2)\rightarrow C(i=3)\rightarrow\cdots$ can be expressed as follows:

$$K_{I,i}\frac{\partial N_{I,i}}{\partial t}=D_I\frac{\partial^2 N_{I,i}}{\partial z_I^2}-V_I\frac{\partial N_{I,i}}{\partial z_I}-\lambda_i K_{I,i}N_{I,i}+\lambda_{i-1}K_{I,i-1}N_{I,i-1} \qquad (1)$$

$$\lambda_0\equiv 0,\ 0\leqq z_I\leqq l_I,\ 0\leqq t,\ i=1,\ 2,\cdots,\ I=1,\ 2,\cdots,$$

where t is time after the start of the release of nuclides from the waste form to geologic media. Subscripts i and I denote a nuclide i and a layer I, respectively. Then, as shown in Fig. 1, z_I is the distance in the I-th layer from the outlet of the $(I-1)$-th layer, and z_1 is that in the first layer from the waste form. The radionuclide concentration $N_{I,i}$ is a function of location z_I and time t. The decay constant, groundwater velocity, dispersion coefficient, retardation factor and layer

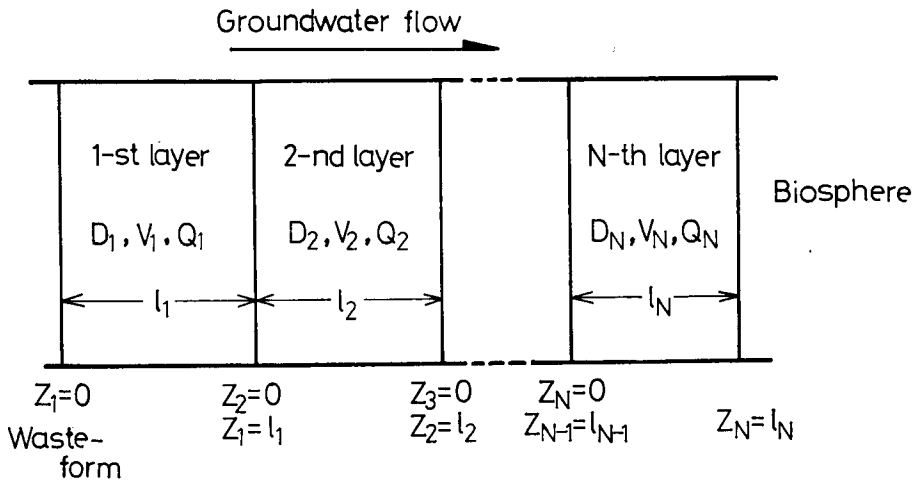

Fig. 1. Schematic representation of a multi-layered migration path.

thickness are denoted by λ, V, D, K and l, respectively.

The initial condition for Eq. (1) is;

$$N_{I,i}(z_I,\ 0)=0,\ I=1,\ 2,\ 3,\cdots \tag{2}$$

The boundary condition at the surface of the waste form (at $z_1=0$) is:

$$N_{1,i}(0,\ t)=\text{arbitrary function}$$
$$=N_{i0}(t) \tag{3}$$

The function $N_{i0}(t)$ should express the time-dependence of the release of a nuclide i from the waste form.

The boundary condition at the interlayer boundary of the $(I-1)$-th layer and I-th layer (at $z_{I-1}=l_{I-1}$ and $z_I=0$) is:

$$N_{I-1,i}(l_{I-1},\ t)=N_{I,i}(0,\ t),\ I=2,\ 3,\cdots \tag{4}$$

The another boundary condition is assumed as follows:

$$N_{I,i}(\infty,\ t)=0 \tag{5}$$

This condition is a substitute for flux conservation at the interlayer boundary. As examined by Hadermann[7], Eq. (5) is a good approximation to flux conservation when an inequality

$$\alpha_I l_I \ll 1 \tag{6}$$

is satisfied, where α_I is dispersivity defined as Eq. (7)[8]:

$$\alpha_I \equiv D_I/V_I \tag{7}$$

By applying the Laplace transformation to Eq. (1) under the conditions of Eqs. (2)-(5), the concentrations can be obtained as follows:

$$N_{I,i}(z_I,\ t)=\mathcal{L}_s^{-1}[\bar{N}_{I,i}(z_I,\ s)] \tag{8}$$

($\mathcal{L}_s^{-1}$: operator of the Laplace inverse transformation)

$$\bar{N}_{I,i}(z_I,\ s)=\sum_{k=1}^{i} C_{i,k}^{(I)}(s)\bar{N}_{k0}(s)P_{I,k}(z_I,\ s) \tag{9}$$

$$P_{I,k}(z_I,\ s)\equiv\exp\left[p_{I,k}(s)z_I+\sum_{K=1}^{I-1}p_{K,k}(s)l_K\right] \tag{10}$$

$$p_{I,i}(s)\equiv\frac{1}{2\alpha_I}\left\{1-\left[1+\frac{4\alpha_I}{v_{I,i}}(s+\lambda_i)\right]^{1/2}\right\} \tag{11}$$

$$C_{i,k}^{(I)}(s)\equiv\begin{cases}\dfrac{h_{i,k}^{(I)}}{s+\beta_{i,k}^{(I)}}C_{i-1,k}^{(I)}(s),\ (I=1,\ 2,\cdots,\ k=1,\ 2,\cdots,\ i-1)\\ 1-\displaystyle\sum_{k'=1}^{i-1}\frac{\bar{N}_{k'0}(s)}{\bar{N}_{i0}(s)}C_{i,k'}^{(1)}(s),\ C_{1,1}^{(1)}(s)\equiv 1\quad (I=1,\ k=i)\\ C_{i,i}^{(I-1)}(s)+\displaystyle\sum_{k'=1}^{i-1}\{C_{i,k}^{(I-1)}(s)-C_{i,k'}^{(I)}(s)\}\\ \qquad\times\exp\left\{\displaystyle\sum_{K=1}^{I-1}[p_{K,k'}(s)-p_{K,i}(s)]l_K\right\}\quad (I=2,\ 3,\cdots,\ k=i)\end{cases} \tag{12}$$

$$h_{i,k}^{(I)}\equiv\frac{\lambda_{i-1}/v_{I,i-1}}{1/v_{I,i}-1/v_{I,k}},\quad v_{I,k}\equiv V_I/K_{I,k},\quad \beta_{i,k}^{(I)}\equiv\frac{v_{I,i}\lambda_k-v_{I,k}\lambda_i}{v_{I,i}-v_{I,k}}$$

As suggested from the recursive from of $C_{i,k}^{(I)}(s)$, the concentration $N_{I,i}(z_I, t)$ is obtained successively from $N_{1,1}(z_1, t)$.

Now we assume the explicit form of the function $N_{i0}(t)$ which depends on the release model adopted. The mathematical treatment to solve Eq. (1) will be much simplified when the Impulse (*or* Accidental)-release model is applied[8]. To make the applicable situations more realistic, however, the Preferential-release model[6] is assumed in this analysis. This release model is a little more general than the Impulse-release model, because one may assume different leachabilities for elements in the Preferential-release model. Furthermore, the solution of Eq. (1) for this release model can be easily transformed into solutions for any of the Exponential-, Band-, Step-, and Impulse-release models[6].

The function $N_{i0}(t)$ corresponding to the Preferential-release model is expressed as:

$$N_{i0}(t)=N_i^0\sum_{j=1}^{i}B'_{i,j}\exp(-\Lambda_j t) \tag{13}$$

where

$$N_i^0\equiv\kappa_i M_1^0/Q_1 \tag{14}$$

$$\Lambda_i\equiv\lambda_i+\kappa_i \tag{15}$$

$$B'_{i,j}\equiv\sum_{m=1}^{j}\frac{(M_m^0/M_1^0)\prod_{r=m}^{i}(\lambda_r/\lambda_i)}{\prod_{\substack{l=m\\ l\neq j}}^{i}(\Lambda_l-\Lambda_j)},\quad B'_{1,1}=1 \tag{16}$$

Here, Q_1 is a volumetric flow rate of groundwater in the 1st layer, M_m^0 is the amount of the nuclide m in the waste form at the beginning of the release ($t=0$), and κ_i is the parameter, called the release rate coefficient in the Preferential-release model. Substitution of Eq. (13) for the function $N_{i0}(t)$ into Eq. (9) leads to:

$$\bar{N}_{I,i}(z_I, s)=\sum_{k=1}^{i}N_k^0 C_{i,k}^{(I)}(s)\sum_{j=1}^{k}\frac{B'_{k,j}}{s+\Lambda_j}P_{I,k}(z_I, s) \tag{17}$$

When one defines the functions as follows

$$E(i, j, k)\equiv\mathcal{L}_s^{-1}\left\{\frac{\exp[p_{1,i}(s)z_1]}{s+\beta_{j,k}^{(1)}}\right\},\quad \beta_{j,0}^{(1)}\equiv\Lambda_j \tag{18}$$

$$F_1(i, j, k, r)\equiv\mathcal{L}_s^{-1}\left\{\frac{\exp[p_{2,i}(s)z_2+p_{1,r}(s)l_1]}{s+\beta_{j,k}^{(1)}}\right\},\quad \beta_{j,0}^{(2)}\equiv\Lambda_j \tag{19}$$

$$F_2(i, j, k, r)\equiv\mathcal{L}_s^{-1}\left\{\frac{\exp[p_{2,i}(s)z_2+p_{1,r}(s)l_1]}{s+\beta_{j,k}^{(2)}}\right\},\quad \beta_{j,0}^{(2)}\equiv\Lambda_j \tag{20}$$

$$F_1(i, j, 0, r)=F_2(i, j, 0, r), \tag{21}$$

the solution $N_{I,i}(z_I, t)$ obtained by the Laplace inverse transformation from Eq. (17) can be formulated linearly with respect to the function $E(i, j, k)$'s for the 1 st layer ($I=1$), and with respect to the functions of $F_1(i, j, k, r)$'s and $F_2(i, j, k, r)$'s for the 2nd layer ($I=2$). The function $E(i, j, k)$ depends on z_1 and t, and

$F_1(i, j, k, r)$ and $F_2(i, j, k, r)$ are functions of z_2 and t. By using these functions, the solutions $N_{I,i}(z_I, t)$ for I(1 or 2) and i(1, 2, 3, or 4) were explicitly presented. For the 1st layer ($I=1$), the solution for the homogeneous layer shown in Ref. (4) can be applied. The solution for the 2nd layer ($I=2$), $N_{2,i}(z_2, t)$, is presented in the Appendix.

III. An Example of Calculation

A decay chain of ^{237}Np-^{233}U-^{229}Th-^{225}Ra was chosen to display the concentration profiles calculated by the solution derived in the preceding section. Neptunium has been regarded as one of the important nuclides that is radiobiologically hazardous. The decay chain is interesting for the study of concentration profiles because (a) the retardation factors of the members vary to a large extent in some types of geologic media, and because (b) the order of magnitude of their retardation factors is sometimes quite different among geologic media. The two-layered geologic media is assumed to be granite follwed by subsoil. Groundwater flows from the first layer of the host rock of granite to the second layer of subsoil bounded by a surface water body.

The hydrogeological parameters used in the calculations are listed in Table I. The retardation factors estimated by KBS[9] for a Swedish granite, and by Burkholder *et al*[10]. for a Western U.S. desert subsoil are adopted. The groundwater velocity is fixed at 1 m/yr for granite and 100 m/yr for subsoil. The dispersion coefficients are assumed to be 1m²/yr for granite and 100 m²/yr for subsoil. The thickness of the 1st layer of granite is fixed at 1,000 m.

Table 1. Hydrogeological Parameters and Data on Radionuclides used in Calculations.

	Groundwater velocity (m/yr)	Dispersion coefficient (m²/yr)
First layer, granite	1	1
Second layer, subsoil	100	100
Thickness of the first layer (m)	1000	

Nuclide	Half-life[a] (yr)	Inventory[b] (Bq/GWe · yr)	$1/K_i(-)$ granite[c],	subsoil[d]	κ_i[e] (1/yr)	ALI[g] (Bq/yr)
^{237}Np	2.14×10^{6}	5.33×10^{11}	4×10^{-3}	1×10^{-2}	6×10^{-8}	3×10^{3}
^{233}U	1.59×10^{5}	0	2×10^{-2}	7×10^{-5}	1×10^{-8}	4×10^{5}
^{229}Th	7.30×10^{3}	0	2×10^{-4}	2×10^{-5}	3×10^{-6}[f]	2×10^{4}
^{225}Ra	4.05×10^{-2}	0	1×10^{-3}	2×10^{-3}	3×10^{-6}[f]	3×10^{5}

a: Ref. (16), *b*: Uranium-fueled 1 GWe PWR, 150 days after discharge from reactor, Ref. (9), *c*: Ref. (12), *d*: Ref. (13), *e*: Ref. (14), *f*: Assumed values, *g*: Ref. (15).

Parameters characteristic to these nuclides are listed also in Table I. The inventory of each radionuclide is based on the radioactivities produced by the operation of nuclear plants of 1,000 GWe · yr[11]. Radionuclides are assumed to be kept in the waste form for 1,000 years. The time-dependence of radioactivity after the discharge of spent fuels is calculated by the so-called Bateman equation[12,13]. In order to simplify the situation, it is also assumed that radionuclides are directly

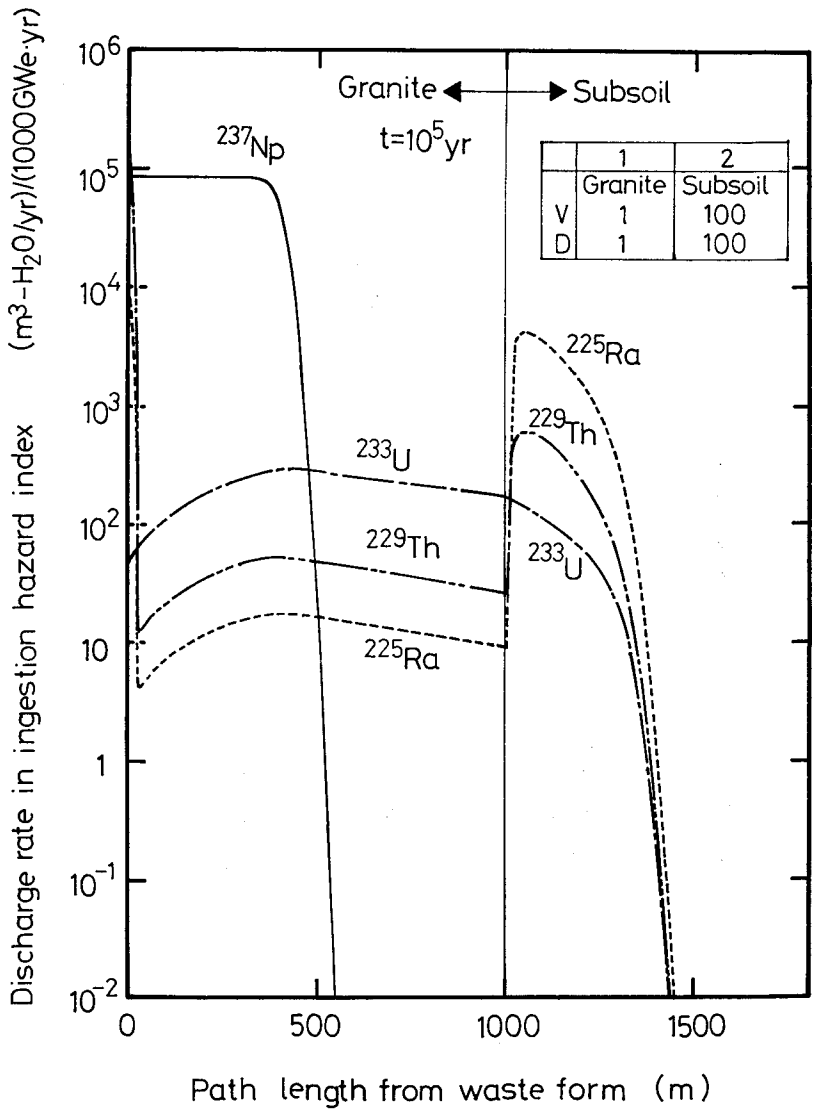

Fig. 2. Annual discharge rates of radionuclides through a two-layered medium: ^{237}Np-^{233}U-^{229}Th-^{225}Ra: 1. granite, 2. subsoil: $t=10^5$yr.

released into a host rock. The release rate coefficient κ_i characterized in the Preferential-release model refers to the solubility-limited dissolution theory given by Chambré *et al.*[14].

The discharge rate of a radionuclide into the biosphere is converted to a potential hazard for man, which is expressed by the so-called Ingestion Hazard Index (IHI) based on ALI (Annual Limit on Intake by Workers).[15] The IHI is defined as

$$(IHI)_i(\mathrm{m^3\text{-}H_2O/yr}) \equiv \frac{R_i}{(ALI)_i/10/0.8} \tag{22}$$

where R_i and $(ALI)_i$ are the annual discharge rate of nuclide i in Bq/yr, and the annual limit of intake by workers for nuclide i in Bq/yr, respectively. In the denominator of Eq. (22), the ALI is divided by the annual water intake 0.8 m³/yr for an adult, and by 10 to obtain a concentration that will limit the annual ingested dose for the public.

The annual discharge rate of each radionuclide migrating through a two-layered medium of granite followed by subsoil is shown in Fig. 2 as a function of the path length from the waste form at $t=10^6$yr after the start of the release. The distributions of Th and its short-lived daughter Ra generated in a repository are limited within a small region of 30 m from a waste form, because their retardation factors are large for the first layer of granite. Thorium and radium present in more than 30 m from the waste form were generated from ^{233}U in granite and are in secular equilibrium. Those two daughters form sharp peaks just outside the interlayer boundary.

Rapid changes in concentrations like those sharp peaks seen in Fig. 2 should be strictly pursued to predict the radionuclide migration. In this sense, those peaks found by an analytical model of this work can serve as benchmark profiles to check the accuracy of the numerical analyses.

The calculations were performed by the computer at The Data Processing Center of Kyoto University.

Appendix

In this Appendix, the solution of $N_{2,i}(z_2, t)$ of Eq. (1) is explicitly given for four-member decay chains. The solution given here can be applied to decay chains whose members have different retardation factors for each and all. The following are defined here, (previously defined as N_i^0 in Eq. (14) is modified):

$$N_i^0 \equiv \begin{cases} \kappa_i M_1^0/Q_1 & i \geqq 1 \\ 0 & i \leqq 0 \end{cases}$$

$$d(i,\ j,\ k,\ r) \equiv [\beta_{i,j}^{(1)} - \beta_{k,r}^{(1)}]^{-1}$$

$$d_2(i,\ j,\ k,\ r)\equiv[\beta^{(2)}_{i,j}-\beta^{(2)}_{k,r}]^{-1}$$
$$d_{12}(i,\ j,\ k,\ r)\equiv[\beta^{(1)}_{i,j}-\beta^{(2)}_{k,r}]^{-1}$$
$$d_{21}(i,\ j,\ k,\ r)\equiv[\beta^{(2)}_{i,j}-\beta^{(1)}_{k,r}]^{-1}$$

With these definitions, the solution for the second layer is as follows:

$$\begin{aligned}
N_{2,i}(z_2,\ t)=&N^0_i\sum_{j=1}^{t}B'_{i,j}F_1(i,\ j,\ 0,\ i)\\
&+N^0_{i-1}h^{(1)}_{i,i-1}\sum_{j=1}^{t-1}B'_{i-1,j}d(i,\ i-1,\ j,\ 0)\{F_1(i,\ j,\ 0,\ i-1)\\
&-F_1(i,\ j,\ 0,\ i)+F_1(i,\ i,\ i-1,\ i)-F_1(i,i,\ i-1,\ i-1)\}\\
&+N^0_{i-1}h^{(2)}_{i,i-1}\sum_{j=1}^{t-1}B'_{i-1,j}d_2(i,\ i-1,\ j,\ 0)\{F_2(i-1,\ j,\ 0,\ i-1)\\
&-F_2(i,\ j,\ 0,\ i-1)+F_2(i,\ i,\ i-1,\ i-1)-F_2(i-1,\ i,\ i-1,\ i-1)\}\\
&+N^0_{i-2}h^{(1)}_{i-1,i-2}h^{(1)}_{i,i-2}\sum_{j=1}^{t-2}B'_{i-2,j}\\
&\times\Big[\begin{array}{l}d(i-1,\ i-2,\ j,\ 0)d(i,\ i-2,\ j,\ 0)\{F_1(i,\ j,\ 0,\ i-2)-F_1(i,j,\ 0,\ i)\}\\
+d(j,\ 0,\ i-1,\ i-2)d(i,\ i-2,\ i-1,\ i-2)\\
\qquad\cdot\{F_1(i,\ i-1,\ i-2,i-2)-F_1(i,\ i-1,\ i-2,\ i)\}\\
+d(j,\ 0,\ i,\ i-2)d(i-1,\ i-2,\ i,\ i-2)\{F_1(i,\ i,\ i-2,\ i-2)\\
\qquad-F_1(i,\ i,\ i-2,\ i)\}\end{array}\Big]\\
&+N^0_{i-2}h^{(1)}_{i-1,i-2}h^{(1)}_{i,i-1}\sum_{j=1}^{t-2}B'_{i-2,j}\\
&\times\Big[\begin{array}{l}d(i-1,\ i-2,\ j,\ 0)d(i,\ i-1,\ j,\ 0)\{F_1(i,\ j,\ 0,\ i)-F_1(i,\ j,\ 0,\ i-1)\}\\
+d(j,\ 0,\ i-1,\ i-2)d(i,\ i-1,\ i-1,\ i-2)\\
\qquad\cdot\{F_1(i,\ i-1,\ i-2,\ i)-F_1(i,\ i-1,\ i-2,\ i)\}\\
+d(j,\ 0,\ i,\ i-1)d(i-1,\ i-2,\ i,\ i-1)\{F_1(i,\ i,\ i-1,\ i)\\
\qquad-F_1(i,\ i,\ i-1,\ i-1)\}\end{array}\Big]\\
&+N^0_{i-2}h^{(1)}_{i-1,i-2}h^{(2)}_{i,i-1}\sum_{j=1}^{t-2}B'_{i-2,j}\\
&\times\Big[\begin{array}{l}d(i-1,\ i-2,\ j,\ 0)d_2(i,\ i-1,\ j,\ 0)\cdot\{F_1(i-1,\ j,\ 0,\ i-2)\\
\qquad-F_1(i-1,j,\ 0,\ i-1)+F_1(i,\ j,\ 0,\ i-1)-F_1(i,\ j,\ 0,\ i-2)\}\\
+d(j,\ 0,\ i-1,\ i-2)d_{21}(i,\ i-1,\ i-1,\ i-2)\\
\qquad\cdot\{F_1(i-1,\ i-1,\ i-2,\ i-2)-F_1(i-1,\ i-1,\ i-2,\ i-1)\\
\qquad+F_1(i,\ i-1,\ i-2,\ i-1)-F_1(i,\ i-1,\ i-2,\ i-2)\}\\
+d_2(j,\ 0,\ i,\ i-1)d_{21}(i-1,\ i-2,\ i,\ i-1)\\
\qquad\cdot\{F_2(i-1,\ i,\ i-1,\ i-2)-F_2(i-1,\ i,\ i-1,\ i-1)\\
\qquad+F_2(i,\ i,\ i-1,\ i-1)-F_2(i,\ i,\ i-1,\ i-2)\end{array}\Big]\\
&+N^0_{i-2}h^{(2)}_{i-1,i-2}h^{(2)}_{i,i-2}\sum_{j=1}^{t-2}B'_{i-2,j}\\
&\Big[\begin{array}{l}d_2(i-1,\ i-2,\ j,\ 0)d_2(i,\ i-2,\ j,0)\cdot\{F_2(i-2,\ j,\ 0,\ i-2)\\
\qquad-F_2(i,\ j,\ 0,\ i-2)\}\end{array}\Big]
\end{aligned}$$

$$\times\left[\begin{array}{l} +d_2(j,\ 0,\ i-1,\ i-2)d_2(i,\ i-2,\ i-1,\ i-2) \\ \qquad \cdot \{F_2(i-2,\ i-1,\ i-2,\ i-2)-F_2(i,\ i-1,\ i-2,\ i-2)\} \\ +d_2(j,\ 0,\ i,\ i-2)d_2(i-1,\ i-2,\ i,\ i-2) \\ \qquad \cdot \{F_2(i-2,\ i,\ i-2,\ i-2)-F_2(i,\ i,i-2,\ i-2)\} \end{array}\right]$$

$$+N^0_{i-3}h^{(2)}_{i-1,i-2}h^{(2)}_{i,i-1}\sum_{j=1}^{i-2}B'_{i-2,j}$$

$$\times\left[\begin{array}{l} d_2(i-1,\ i-2,\ j,\ 0)d_2(i,\ i-1,\ j,\ 0) \\ \qquad \cdot \{F_2(i,\ j,\ 0,\ i-2)-F_2(i-1,\ j,\ 0,\ i-2)\} \\ +d_2(j,\ 0,\ i-1,\ i-2)d_2(i,\ i-1,\ i-1,\ i-2) \\ \qquad \cdot \{F_2(i,\ i-1,\ i-2,\ i-2)-F_2(i-1,\ i-1,\ i-2,\ i-2)\} \\ +d_2(j,\ 0,\ i,\ i-1)d_2(i-1,\ i-2,\ i,\ i-1) \\ \qquad \cdot \{F_2(i,\ i,\ i-1,\ i-2)-F_2(i-1,\ i,\ i-1,\ i-2)\} \end{array}\right]$$

$$+N^0_{i-3}h^{(1)}_{2,1}h^{(1)}_{3,1}h^{(1)}_{4,1}$$

$$\times\left[\begin{array}{l} d(2,\ 1,\ 1,\ 0)d(3,\ 1,\ 1,\ 0)d(4,\ 1,\ 1,\ 0)\{F_1(4,\ 1,\ 0,\ 1)-F_1(4,\ 1,\ 0,\ 4)\} \\ +d(1,\ 0,\ 2,\ 1)d(3,\ 1,\ 2,\ 1)d(4,\ 1,\ 2,\ 1)\{F_1(4,\ 2,\ 1,\ 1)-F_1(4,\ 2,\ 1,\ 4)\} \\ +d(1,\ 0,\ 3,\ 1)d(2,\ 1,\ 3,\ 1)d(4,\ 1,\ 3,\ 1)\{F_1(4,\ 3,\ 1,\ 1)-F_1(4,\ 3,\ 1,\ 4)\} \\ +d(1,\ 0,\ 4,\ 1)d(2,\ 1,\ 4,\ 1)d(3,\ 1,\ 4,\ 1)\{F_1(4,\ 4,\ 1,\ 1)-F_1(4,\ 4,\ 1,\ 4)\} \end{array}\right]$$

$$+N^0_{i-3}h^{(1)}_{2,1}h^{(1)}_{3,1}h^{(1)}_{4,3}$$

$$\times\left[\begin{array}{l} d(2,\ 1,\ 1,\ 0)d(3,\ 1,\ 1,\ 0)d(4,\ 3,\ 1,\ 0)\{F_1(4,\ 1,\ 0,\ 4)-F_1(4,\ 1,\ 0,\ 3)\} \\ +d(1,\ 0,\ 2,\ 1)d(3,\ 1,\ 2,\ 1)d(4,\ 3,\ 2,\ 1)\{F_1(4,\ 2,\ 1,\ 4)-F_1(4,\ 2,\ 1,\ 3)\} \\ +d(1,\ 0,\ 3,\ 1)d(2,\ 1,\ 3,\ 1)d(4,\ 3,\ 3,\ 1)\{F_1(4,\ 3,\ 1,\ 4)-F_1(4,\ 3,\ 1,\ 3)\} \\ +d(1,\ 0,\ 4,\ 3)d(2,\ 1,\ 4,\ 3)d(3,\ 1,\ 4,\ 3)\{F_1(4,\ 4,\ 3,\ 4)-F_1(4,\ 4,\ 3,\ 3)\} \end{array}\right]$$

$$+N^0_{i-3}h^{(1)}_{2,1}h^{(1)}_{3,2}h^{(1)}_{4,2}$$

$$\times\left[\begin{array}{l} d(2,\ 1,\ 1,\ 0)d(3,\ 1,\ 1,\ 0)d(4,\ 3,\ 1,\ 0)\{F_1(4,\ 1,\ 0,\ 4)-F_1(4,\ 1,\ 0,\ 2)\} \\ +d(1,\ 0,\ 2,\ 1)d(3,\ 2,\ 2,\ 1)d(4,\ 2,\ 2,\ 1)\{F_1(4,\ 2,\ 1,\ 4)-F_1(4,\ 2,\ 1,\ 2)\} \\ +d(1,\ 0,\ 3,\ 2)d(2,\ 1,\ 3,\ 2)d(4,\ 2,\ 3,\ 2)\{F_1(4,\ 3,\ 2,\ 4)-F_1(4,\ 3,\ 2,\ 2)\} \\ +d(1,\ 0,\ 4,\ 2)d(2,\ 1,\ 4,\ 2)d(3,\ 2,\ 4,\ 2)\{F_1(4,\ 4,\ 2,\ 4)-F_1(4,\ 4,\ 2,\ 2)\} \end{array}\right]$$

$$+N^0_{i-3}h^{(1)}_{2,1}h^{(1)}_{3,2}h^{(1)}_{4,3}$$

$$\times\left[\begin{array}{l} d(2,\ 1,\ 1,\ 0)d(3,\ 2,\ 1,\ 0)d(4,\ 3,\ 1,\ 0)\{F_1(4,\ 1,\ 0,\ 3)-F_1(4,\ 1,\ 0,\ 4)\} \\ +d(1,\ 0,\ 2,\ 1)d(3,\ 2,\ 2,\ 1)d(4,\ 3,\ 2,\ 1)\{F_1(4,\ 2,\ 1,\ 3)-F_1(4,\ 2,\ 1,\ 4)\} \\ +d(1,\ 0,\ 3,\ 2)d(2,\ 1,\ 3,\ 2)d(4,\ 3,\ 3,\ 2)\{F_1(4,\ 3,\ 2,\ 3)-F_1(4,\ 3,\ 2,\ 4)\} \\ +d(1,\ 0,\ 4,\ 3)d(2,\ 1,\ 4,\ 3)d(3,\ 2,\ 4,\ 3)\{F_1(4,\ 4,\ 3,\ 3)-F_1(4,\ 4,\ 3,\ 4)\} \end{array}\right]$$

$$+N^0_{i-3}h^{(1)}_{2,1}h^{(1)}_{3,1}h^{(2)}_{4,3}$$

$$\times\left[\begin{array}{l} d(2,\ 1,\ 1,\ 0)d(3,\ 1,\ 1,\ 0)d_2(4,\ 3,\ 1,\ 0)\cdot\{F_1(3,\ 1,\ 0,\ 1) \\ \qquad -F_1(3,\ 1,\ 0,\ 3)+F_1(4,\ 1,\ 0,\ 3)-F_1(4,\ 1,\ 0,\ 1)\} \\ +d(1,\ 0,\ 2,\ 1)d(3,\ 1,\ 2,\ 1)d_{21}(4,3,\ 2,\ 1)\cdot\{F_1(3,\ 2,\ 1,\ 1) \\ \qquad -F_1(3,\ 2,\ 1,\ 3)+F_1(4,\ 2,\ 1,\ 3)-F_1(4,\ 2,\ 1,\ 1)\} \\ +d(1,\ 0,\ 3,\ 1)d(2,\ 1,\ 3,\ 1)d_{21}(4,\ 3,\ 3,\ 1)\cdot\{F_1(3,\ 3,\ 1,\ 1) \end{array}\right]$$

$$
\left[\begin{array}{l}
\qquad -F_1(3,\ 3,\ 1,\ 3)+F_1(4,\ 3,\ 1,\ 3)-F_1(4,\ 3,\ 1,\ 1)\} \\
+d_2(1,\ 0,\ 4,\ 3)d_{12}(2,\ 1,\ 4,\ 3)d_{12}(3,\ 1,\ 4,\ 3)\cdot\{F_2(3,\ 4,\ 3,\ 1) \\
\qquad -F_2(3,\ 4,\ 3,\ 3)+F_2(4,\ 4,\ 3,\ 3)-F_2(4,\ 4,\ 3,\ 1)\}
\end{array}\right]
$$

$$
+N^0_{i-3}h^{(1)}_{2,1}h^{(1)}_{3,2}h^{(2)}_{4,3}
$$

$$
\times\left[\begin{array}{l}
d(2,\ 1,\ 1,\ 0)d(3,\ 2,\ 1,\ 0)d_2(4,\ 3,\ 1,\ 0)\cdot\{F_1(3,\ 1,\ 0,\ 3) \\
\qquad -F_1(3,\ 1,\ 0,\ 2)+F_1(4,\ 1,\ 0,\ 2)-F_1(4,\ 1,\ 0,\ 3)\} \\
+d(1,\ 0,\ 2,\ 1)d(3,\ 2,\ 2,\ 1)d_{21}(4,\ 3,\ 2,\ 1)\cdot\{F_1(3,\ 2,\ 1,3) \\
\qquad -F_1(3,\ 2,\ 1,\ 2)+F_1(4,\ 2,\ 1,\ 2)-F_1(4,\ 2,\ 1,\ 3)\} \\
+d(1,\ 0,\ 3,\ 2)d(2,\ 1,\ 3,\ 2)d_{21}(4,\ 3,\ 3,\ 2)\cdot\{F_1(3,\ 3,\ 2,\ 3) \\
\qquad -F_1(3,\ 3,\ 2,\ 2)+F_1(4,\ 3,\ 2,\ 2)-F_1(4,\ 3,\ 2,\ 3)\} \\
+d_2(1,\ 0,\ 4,\ 3)d_{12}(2,\ 1,\ 4,\ 3)d_{12}(3,\ 2,\ 4,\ 3)\cdot\{F_2(3,\ 4,\ 3,\ 3) \\
\qquad -F_2(3,\ 4,\ 3,\ 2)+F_2(4,\ 4,\ 3,\ 2)-F_2(4,\ 4,\ 3,\ 3)\}
\end{array}\right]
$$

$$
+N^0_{i-3}h^{(1)}_{2,1}h^{(2)}_{3,2}h^{(2)}_{4,2}
$$

$$
\times\left[\begin{array}{l}
d(2,\ 1,\ 1,\ 0)d_2(3,\ 2,\ 1,\ 0)d_2(4,\ 2,\ 1,\ 0)\cdot\{F_1(2,\ 1,\ 0,\ 1) \\
\qquad -F_1(2,\ 1,\ 0,\ 2)+F_1(4,\ 1,\ 0,\ 2)-F_1(4,\ 1,\ 0,\ 1)\} \\
+d(1,\ 0,\ 2,\ 1)d_{21}(3,\ 2,\ 2,\ 1)d_{21}(4,\ 2,\ 2,\ 1)\cdot\{F_1(2,\ 2,\ 1,\ 1) \\
\qquad -F_1(2,\ 2,\ 1,\ 2)+F_1(4,\ 2,\ 1,\ 2)-F_1(4,\ 2,\ 1,\ 1)\} \\
+d_2(1,\ 0,\ 3,\ 2)d_{12}(2,\ 1,\ 3,\ 2)d_2(4,\ 2,\ 3,\ 2)\cdot\{F_2(2,\ 3,\ 2,\ 1) \\
\qquad -F_2(2,\ 3,\ 2,\ 2)+F_2(4,\ 3,\ 2,\ 2)-F_2(4,\ 3,\ 2,\ 1)\} \\
+d_2(1,\ 0,\ 4,\ 2)d_{12}(2,\ 1,\ 4,\ 2)d_2(3,\ 2,\ 4,\ 2)\cdot\{F_2(2,\ 4,\ 2,\ 1) \\
\qquad -F_2(2,\ 4,\ 2,\ 2)+F_2(4,\ 4,\ 2,\ 2)-F_2(4,\ 4,\ 2,\ 1)\}
\end{array}\right]
$$

$$
+N^0_{i-3}h^{(1)}_{2,1}h^{(2)}_{3,2}h^{(2)}_{4,3}
$$

$$
\times\left[\begin{array}{l}
d(2,\ 1,\ 1,\ 0)d_2(3,\ 2,\ 1,\ 0)d_2(4,\ 3,\ 1,\ 0)\cdot\{F_1(4,\ 1,\ 0,\ 1) \\
\qquad -F_1(4,\ 1,\ 0,\ 2)+F_1(3,\ 1,\ 0,\ 2)-F_1(3,\ 1,\ 0,\ 1)\} \\
+d(1,\ 0,\ 2,\ 1)d_{21}(3,\ 2,\ 2,\ 1)d_{21}(4,\ 3,\ 2,\ 1)\cdot\{F_1(4,\ 2,\ 1,\ 1) \\
\qquad -F_1(4,\ 2,\ 1,\ 2)+F_1(3,\ 2,\ 1,\ 2)-F_1(3,\ 2,\ 1,\ 1)\} \\
+d_2(1,\ 0,\ 3,\ 2)d_{12}(2,\ 1,\ 3,\ 2)d_2(4,\ 3,\ 3,\ 2)\cdot\{F_2(4,\ 3,\ 2,\ 1) \\
\qquad -F_2(4,\ 3,\ 2,\ 2)+F_2(3,\ 3,\ 2,\ 2)-F_2(3,\ 3,\ 2,\ 1)\} \\
+d_2(1,\ 0,\ 4,\ 3)d_{12}(2,\ 1,\ 4,\ 3)d_2(3,\ 2,\ 4,\ 3)\cdot\{F_2(4,\ 4,\ 3,\ 1) \\
\qquad +F_2(4,\ 4,\ 3,\ 2)+F_2(3,\ 4,\ 3,\ 2)-F_2(3,\ 4,\ 3,\ 1)\}
\end{array}\right]
$$

$$
+N^0_{i-3}h^{(2)}_{2,1}h^{(2)}_{3,1}h^{(2)}_{4,1}
$$

$$
\times\left[\begin{array}{l}
d_2(2,\ 1,\ 1,\ 0)d_2(3,\ 1,\ 1,\ 0)d_2(4,\ 1,\ 1,\ 0) \\
\qquad\qquad\qquad\qquad\qquad\qquad\cdot\{F_2(1,\ 1,\ 0,\ 1)-F_2(4,\ 1,\ 0,\ 1)\} \\
+d_2(1,\ 0,\ 2,\ 1)d_2(3,\ 1,\ 2,\ 1)d_2(4,\ 1,\ 2,\ 1) \\
\qquad\qquad\qquad\qquad\qquad\qquad\cdot\{F_2(1,\ 2,\ 1,\ 1)-F_2(4,\ 2,\ 1,\ 1)\} \\
+d_2(1,\ 0,\ 3,\ 1)d_2(2,\ 1,\ 3,\ 1)d_2(4,\ 1,\ 3,\ 1) \\
\qquad\qquad\qquad\qquad\qquad\qquad\cdot\{F_2(1,\ 3,\ 1,\ 1)-F_2(4,\ 3,\ 1,\ 1)\} \\
+d_2(1,\ 0,\ 4,\ 1)d_2(2,\ 1,\ 4,\ 1)d_2(3,\ 1,\ 4,\ 1)
\end{array}\right]
$$

$$\cdot \{F_2(1,\ 4,\ 1,\ 1)-F_2(4,\ 4,\ 1,\ 1)\}$$

$$+N^0_{i-3}h^{(2)}_{2,1}h^{(2)}_{3,1}h^{(2)}_{4,3}$$

$$\begin{aligned}
&d_2(2,\ 1,\ 1,\ 0)d_2(3,\ 1,\ 1,\ 0)d_2(4,\ 3,\ 1,\ 0)\\
&\qquad\cdot \{F_2(4,\ 1,\ 0,\ 1)-F_2(3,\ 1,\ 0,\ 1)\}\\
&+d_2(1,\ 0,\ 2,\ 1)d_2(3,\ 1,\ 2,\ 1)d_2(4,\ 3,\ 2,\ 1)\\
&\qquad\cdot \{F_2(4,\ 2,\ 1,\ 1)-F_2(3,\ 2,\ 1,\ 1)\}\\
&+d_2(1,\ 0,\ 3,\ 1)d_2(2,\ 1,\ 3,\ 1)d_2(4,\ 3,\ 3,\ 1)\\
&\qquad\cdot \{F_2(4,\ 3,\ 1,\ 1)-F_2(3,\ 3,\ 1,\ 1)\}\\
&+d_2(1,\ 0,\ 4,\ 3)d_2(2,\ 1,\ 4,\ 3)d_2(3,\ 1,\ 4,\ 3)\\
&\qquad\cdot \{F_2(4,\ 4,\ 3,\ 1)-F_2(3,\ 4,\ 3,\ 1)\}
\end{aligned}$$

$$+N^0_{i-3}h^{(2)}_{2,1}h^{(2)}_{3,2}h^{(2)}_{4,2}$$

$$\begin{aligned}
&d_2(2,\ 1,\ 1,\ 0)d_2(3,\ 2,\ 1,\ 0)d_2(4,\ 2,\ 1,\ 0)\\
&\qquad\cdot \{F_2(4,\ 1,\ 0,\ 1)-F_2(2,\ 1,\ 0,\ 1)\}\\
&+d_2(1,\ 0,\ 2,\ 1)d_2(3,\ 2,\ 2,\ 1)d_2(4,\ 2,\ 2,\ 1)\\
&\qquad\cdot \{F_2(4,\ 2,\ 1,\ 1)-F_2(2,\ 2,\ 1,\ 1)\}\\
&+d_2(1,\ 0,\ 3,\ 2)d_2(2,\ 1,\ 3,\ 2)d_2(4,\ 2,\ 3,\ 2)\\
&\qquad\cdot \{F_2(4,\ 3,\ 2,\ 1)-F_2(2,\ 3,\ 2,\ 1)\}\\
&+d_2(1,\ 0,\ 4,\ 2)d_2(2,\ 1,\ 4,\ 2)d_2(3,\ 2,\ 4,\ 2)\\
&\qquad\cdot \{F_2(4,\ 4,\ 2,\ 1)-F_2(2,\ 4,\ 2,\ 1)\}
\end{aligned}$$

$$+N^0_{i-3}h^{(2)}_{2,1}h^{(2)}_{3,2}h^{(2)}_{4,3}$$

$$\begin{aligned}
&d_2(2,\ 1,\ 1,\ 0)d_2(3,\ 2,\ 1,\ 0)d_2(4,\ 3,\ 1,\ 0)\\
&\qquad\cdot \{F_2(3,\ 1,\ 0,\ 1)-F_2(4,\ 1,\ 0,\ 1)\}\\
&+d_2(1,\ 0,\ 2,\ 1)d_2(3,\ 2,\ 2,\ 1)d_2(4,\ 3,\ 2,\ 1)\\
&\qquad\cdot \{F_2(3,\ 2,\ 1,\ 1)-F_2(4,\ 2,\ 1,\ 1)\}\\
&+d_2(1,\ 0,\ 3,\ 2)d_2(2,\ 1,\ 3,\ 2)d_2(4,\ 3,\ 3,\ 2)\\
&\qquad\cdot \{F_2(3,\ 3,\ 2,\ 1)-F_2(4,\ 3,\ 2,\ 1)\}\\
&+d_2(1,\ 0,\ 4,\ 3)d_2(2,\ 1,\ 4,\ 3)d_2(3,\ 2,\ 4,\ 3)\\
&\qquad\cdot \{F_2(3,\ 4,\ 3,\ 1)-F_2(4,\ 4,\ 3,\ 1)\}
\end{aligned}$$

The functions $F_1(i,\ j,\ k,\ r)$ and $F_2(i,\ j,\ k,\ r)$ defined as Eqs. (17) and (18), respectively, can be expanded to analytical forms by similar mathematical procedures applied to $E(i,\ j,\ k)$ in Ref. (4).

$$\begin{aligned}
F_1(i,\ j,\ k,\ r) &\equiv \mathcal{L}_s^{-1}\left\{\frac{\exp[p_{2,i}(s)z_2+p_{1,r}(s)z_1]}{s+\beta^{(1)}_{j,k}}\right\}\\
&=\frac{4}{\pi}\exp\left(\frac{z_2}{2\alpha_2}+\frac{l_1}{2\alpha_1}-\beta^{(1)}_{j,k}t\right)\\
&\quad\times\int_{a_{1,r}l_1/2\sqrt{t}}^{\eta=\infty}\int_{a_{2,i}z_2/2\sqrt{t}}^{\xi=\infty}\exp\{-[\xi^2+\gamma^{(2)}_{ijk}(a_{2,i}z_2/2\xi)^2]\}\\
&\quad\times\exp\{-[\eta^2+\gamma^{(1)}_{rjk}(a_{1,r}l_1/2\eta)^2]\}d\xi d\eta \qquad \text{(A-1)}
\end{aligned}$$

where

$$a_{I,i} \equiv 1/\sqrt{\alpha_I v_{I,i}} = \sqrt{K_{I,i}/D_I}$$

$$\gamma_{ijk}^{(I)} \equiv \lambda_i + v_{I,i}/4\alpha_I - \beta_{j,k}^{(1)}$$

$$t' \equiv t - (a_{1,r} l_1/2\eta)^2$$

When $\gamma_{ijk}^{(2)} \geqq 0$, Eq. (A-1) can be transformed into the following form:

$$F_1(i,\ j,\ k,\ r) = \frac{1}{\sqrt{\pi}} \exp\left(\frac{z_2}{2\alpha_2} + \frac{l_1}{2\alpha_1} - \beta_{j,k}^{(1)} t\right)$$
$$\times \int_{a_{1,r} l_1/2\sqrt{t}}^{\eta=\infty} \exp\{-[\eta^2 + \gamma_{rjk}^{(1)} (a_{1,r} l_1/2\eta)^2]\}$$
$$\times [\exp(a_{2,i} z_2 \sqrt{\gamma_{ijk}^{(2)}})\, erfc(a_{2,i} z_2/2\sqrt{t'} + \sqrt{\gamma_{ijk}^{(2)} t'})$$
$$+ \exp(-a_{2,i} z_2 \sqrt{\gamma_{ijk}^{(2)}})\, erfc(a_{2,i} z_2/2\sqrt{t'} - \sqrt{\gamma_{ijk}^{(2)} t'})]\, d\eta \qquad \text{(A-2)}$$

where

$$erfc(x) = \frac{2}{\sqrt{\pi}} \int_x^\infty e^{-u^2} du$$

In Eq. (A-2), the remaining single integration must be calculated numerically. Numerical double integration has to be applied when $\gamma_{ijk}^{(2)}$ is negative in Eq. (A-1).

A similar form of the function $F_2(i,\ j,\ k,\ r)$ can easily be obtained by the substitution of $\beta_{j,k}^{(2)}$ for $\beta_{j,k}^{(1)}$ in the function $F_1(i,\ j,\ k,\ r)$.

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