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A Nuclide Decay Chain Transport Model by the Method of Characteristics

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Abstract

The nuclide transport in the one-dimensional porous medium is considered as a first step in developing a decay chain transport in multidimensional inhomogeneous media. A method of solving conventional advection-dispersion equation with decay chain of arbitrary length by using the method of characteristics (MOC) is introduced. In specific cases where the advection are dominant rather than dispersion, the method is known to be useful : one of the most distinctive advantages in applying the model is that the MOC minimizes the numerical dispersion, which is distinguished in such common numerical schemes as finite element method and finite difference method. The suggested model is considered to be effective through several illustrations for the case that decay chain of arbitrary length is involved during transport, which is difficult to solve by standard numerical solutions if the medium becomes more complicated.

1. Introduction

For most practical problems concerning nuclide transport in the medium around the high-level radioactive waste repository, the conventional advection-dispersion equation with decay chain terms is usually considered. In such case, it is very common to obtain the solution for breakthrough curve for nuclide concentration numerically. However many such numerical approaches are usually suffering from problems associated with numerical dispersion and spatial oscillation near the concentration front. Furthermore, application of restrictive criteria for the spatial and temporal discretization results in fine grid size and small time steps making the solution impractical in applying to real scale transport simulation.

In many cases the advection is considered to be the dominant mechanism rather than the dispersion for the transport in natural barrier around the repository, which facilitates the method of characteristics (MOC) in solving the advection-dispersion equation. Since Garder et al. (1964) implemented MOC in the transport problem by utilizing particle tracking to simulate the characteristics line in conjunction with finite difference method, many extended works have been carried out. (Konikow and Bredehoef, 1978 ; Reddel and Sunada, 1970 ; Khaleel and Reddel, 1986) Also, recently, Goode and Konikow (1989) presented the MOC model including single decay and sorption phenomena and Zheng (1993) further extended to three dimensional solute transport.

In this paper as another extended work, an approach to implementation of decay chain of arbitrary length in one-dimensional advection-dispersion equation by utilizing MOC is discussed.

2. Model

The governing equations of advection-dispersion with decay chain assuming one-dimensional semi-finite medium are

$$R_1\frac{\partial c_1}{\partial t}+v\frac{\partial c_1}{\partial x}+\lambda_1 R_1 c_1 = D_1\frac{\partial^2 c_1}{\partial x^2}$$

$$R_2\frac{\partial c_2}{\partial t}+v\frac{\partial c_2}{\partial x}+\lambda_2 R_2 c_2 = D_2\frac{\partial^2 c_2}{\partial x^2}+\lambda_1 R_1 c_1$$

..... (1)

$$R_l\frac{\partial c_l}{\partial t}+v\frac{\partial c_l}{\partial x}+\lambda_l R_l c_l = D_l\frac{\partial^2 c_l}{\partial x^2}+\lambda_{l-1} R_{l-1} c_{l-1}$$

.....

where c_l is the concentration of nuclide component l, λ_l and R_l are the first order decay constant and the retardation coefficient of component l in the one-dimensional medium. All other symbols have their conventional meaning. Above Equations are subject to the following side conditions

$$c_l(x,0)=0 \quad (2a)$$

$$c_l(\infty,t)=0 \quad (2b)$$

$$c_l(0,t)=\sum_{m=1}^{l} B_{lm}e^{-\lambda_m t},\ l=1,2,\ldots \quad (2c)$$

where B_{lm} is the coefficient of the Bateman equation represented as

$$B_{lm}=\sum_{k=1}^{m}\frac{c_k^0}{\lambda_l}\frac{\prod_{r\neq k}^{l}\lambda_r}{\prod_{\substack{n=k\\ n\neq m}}^{l}(\lambda_n-\lambda_m)}$$

and c_k^0 is the initial concentration of the kth member.

In this study no flow boundary condition at the outlet of the system which is a special case of a constant flux boundary condition is exclusively considered. However, different kind of boundary conditions can be incorporated into the model considered here. With no flow boundary condition, the transport across the outlet boundary, i.e., out of the final node of the system is precluded.

In Eq.(1) ($\partial c_l/\partial t$ represents an Eulerian expression representing the rate of change in nuclide concentration at the fixed point in space, which also can be expressed as a Lagrangian form introducing total derivative Dc_l/Dt, which implicates the rate of change in nuclide concentration c_l along the pathline of the nuclide particle and also can be interpreted as a characteristic curve of the velocity field.

Therefore for nuclide l, Eq.(1) can be transformed with $\frac{Dc_l}{Dt}=\frac{\partial c_l}{\partial t}+\frac{v}{R_l}\frac{\partial c_l}{\partial x}$

$$\frac{Dc_l}{Dt}=\frac{D_l}{R_l}\frac{\partial^2 c_l}{\partial x^2}-\lambda_l c_l+\lambda_{l-1}c_{l-1} \quad (3)$$

where $\lambda=0$ when $l=1$.

Above Eq.(3) is approximated again through the finite difference

$$\frac{Dc_l}{Dt}\cong\frac{c_l^{t+\Delta t}-c_l^{*}}{\Delta t} \quad (4)$$

where $c_l^{t+\Delta t}$ is the average nuclide concentration at time $(t+\Delta t)$ and c_l^{*} is the average nuclide concentration in the specific grid block at time t due to advection alone. Let asterisk(*) mean this *intermediate* time level.

Then Eq.(3) is expressed as

$$c_l^{t+\Delta t}\cong c_l^{*}+\Delta t\cdot\left\{\frac{D_l}{R_l}\frac{\partial^2 c_l}{\partial x^2}-\lambda_l c_l+\lambda_{l-1}c_{l-1}\right\} \quad (5)$$

Meanwhile Eq.(1) has one characteristic as follows :

$$\frac{dx}{dt}=\frac{v}{R_l} \quad (6)$$

Along these characteristic curves, Eq.(1) is reduced to R.H.S. of Eq.(3) and then Eq.(1) can be approximated as a coupled form of Eq.(5) and Eq.(6). This is accomplished by adopting a finite difference grid. First, Eq. (6) is solved by introducing a set of moving particles in cell of the finite difference grid block and then tracking the movement of these particles in turn. By utilizing the information about these particles

for their position and concentration associated with it, the concentration at each grid due to advective transport can be evaluated. Following this, Eq. (5) is used to further evaluate changes in each nodal concentration resulting from hydrodynamic dispersion and the sink, *i.e.*, radioactive decay chain.

As soon as a set of moving particles with initial concentration having their own coordinates in the one-dimensional x direction are introduced, they will relocate in the new time level according to the following first-order Euler algorithm for pth moving particle :

$$x_{p,l}^{t+\Delta t} = x_p^t + \Delta t \cdot \frac{v}{R_l} \tag{7}$$

Then, when all the moving particles have been relocated, each grid block is temporarily assigned a concentration $c_l^{t+\Delta t}$ by taking the average $c_p^{t+\Delta t}$ of the concentrations of all the moving particles lying inside the grid block at time level $(t+\Delta t)$. This completes the transport due to advection only. Next, Δc which represents the additional change of the concentration owing to both dispersion and radioactive decay is calculated. Once Δc is evaluated, then each moving particle is assigned a modified concentration according to following Eq. (8) :

$$c_{p,l}^{t+\Delta t} = c_{p,l}^t + \Delta c_{p,l}^t \tag{8}$$

and the nuclide concentration in the specific grid is then calculated as

$$c_l^{t+\Delta t} = c_l^{t^*} + \Delta c_l^{t+\Delta t} \tag{9}$$

To evaluate the additional term for concentration changes only due to hydrodynamic dispersion, Δc in Eq.(8), which is identified again as the last term of Eq.(5), is approximated by utilizing the explicit centered finite difference scheme with temporary exclusion of the decay chain term for simplicity in the form as

$$\Delta c_{l,i}^{t+\Delta t} \cong \frac{D_l \cdot \Delta t}{R_l \cdot (\Delta x)^2}\left(c_{l,i-1}^t - 2c_{l,i}^t + c_{l,i+1}^t\right) \tag{10}$$

Also in order to achieve numerical stability using time stepping difference scheme, the size of the temporal step Δt must be chosen properly to satisfy certain stability criteria. To this end usual stability check corresponding to criterion normally used in the explicit finite difference scheme is made. According to such criterion,

$$\Delta t \le \min\left\{\frac{R_l(\Delta x)^2}{2D_l}\right\} \quad \text{for all grids} \tag{11}$$

In addition, another criterion related to the relocation of moving particles in accordance with Eq. (7) should be made by utilizing familiar Courant-Friedrichs condition for the finite difference scheme of advective transport equation as follows (Huyakorn and Pinder, 1983):

$$\frac{v \cdot \Delta t}{R_l \cdot \Delta x} \le 1 \tag{12}$$

In the case of presence of decay term as shown in Eq.(1), decay chain can be simulated by considering the decay as change of chemical character ; the character of particles representing the parent nuclide is not changed during their decay, whereas the character of the moving particles representing the daughters has been changed while they are generated due to decay of the parent and keep on transporting. The number of daughter particles generated accordingly are equal to the number of parent decayed. After generation of the daughter it follows the same destiny to their parent.

Suppose that a moving particle representing the parent (l-1) is released at time 0 and reaches $x=x_1$ during Δt. Then the particle number of parent will be reduced to

$$N_{l-1}(x_1, \Delta t) = N_{l-1}^0 e^{-\lambda_{l-1}\Delta t} \tag{13}$$

Naturally the particle number of daughter nuclide will then be generated according to

$$N_l(x_1, \Delta t) = N_{l-1}^0\left(1 - e^{-\lambda_{l-1}\Delta t}\right) \tag{14}$$

On account of the linearity of Eq.(1) the solutions for the concentration of a lth member could be given by

$$
\begin{aligned}
c_1(x,t) &= c_1^{(1)}(x,t) \\
c_2(x,t) &= c_2^{(1)}(x,t) + c_2^{(2)}(x,t) \\
&\cdots\cdots\cdots\cdots \\
c_l(x,t) &= c_l^{(1)}(x,t) + c_l^{(2)}(x,t) \\
&\cdots\cdots\cdots\cdots
\end{aligned}
\qquad (15)
$$

where $c_l^{(1)}(x, t)$ and $c_l^{(2)}(x, t)$ are precursor contributions stemming from $c_l(x, t)$ and $c_{l-1}(1-e^{-\lambda_{l-1}\Delta t})$, respectively.

The algorithm utilized is graphically shown in Fig.1.

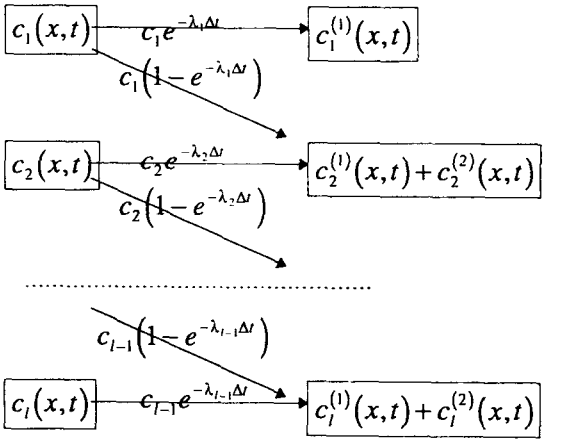

Fig. 1. Algorithm for Chain Decay Calculation.

3. Results and Discussion

3.1. Numerical Implementation

The calculation is carried out by following the algorithm which is briefly described as : (1) locating of moving particles in each grid block i ; (2) relocating the moving particles according to the advective transport ; (3) calculating temporary concentration due to advection only by averaging the concentrations of moving particles ; (4) updating the concentration by considering dispersion ; and finally (5) linearly superpositioning the concentration for chain decay and going to next time step.

3.2. Sensitivity Study

Since it is very difficult to obtain the optimum criteria theoretically by which the validity of numerical calculation can be retained, experimental calculations with several numerical parameters are of necessity. To this end in this study sensitivity to two numerical parameters such as grid size Δx and number of moving particles per each grid block are carried out with fixed time step size for both cases. Even though the results are not presented in this study, the sensitivity to time step size shows no great dependency on the results. This could be explained in following way: advection, which is dominant mechanism of transport, is simulated by Lagrangian particle tracking method based on moving coordinates whereas only dispersion and decay terms are based on finite difference scheme. These parameters are chosen in the limit of the general criteria for finite difference scheme as described in Eqs.(11) and (12).

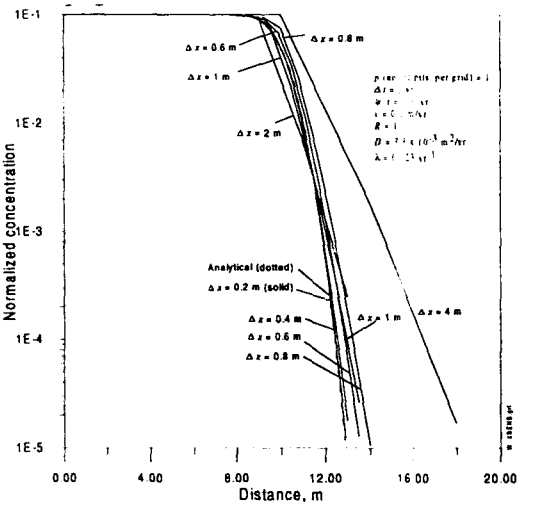

Fig. 2. Sensitivities to Spatial Grid Sizes.

First of all, to show the effect of grid size on the difference with analytical solution provided by Lung (1986), several calculations are made using several different values for grid size when other parameters remains fixed. The parameter values used are : v = 0.1m/yr, $D = 3.3\times10^{-3}\mathrm{m}^2/\mathrm{yr}$, $\lambda = 0{,}023^{-1}\mathrm{yr}$, $\Delta t = 1.0$,

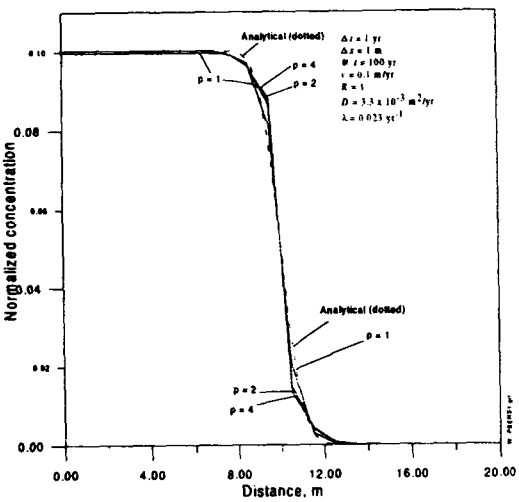

Fig. 3. Sensitivities to the Number of Moving Particles with Δx=1m.

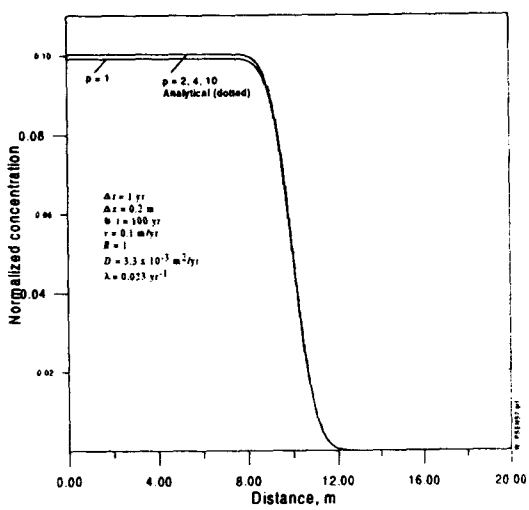

Fig. 4. Sensitivities to the Number of Moving Particles with Δx=0.2m.

and Δx=0.2 to 4. The resulting profiles of these runs for fixed temporal step size are shown in Fig. 2. As shown in the figure, which is enlarged locally for easily understanding, and as expected, the difference seems to get smaller with grid size. Accordingly, generally speaking, for case of Δx<2.0m, the difference could be believed to be negligible.

Figs. 3 and 4 show the concentration profiles at the same time lapsed for the same parameter values listed in Table 1, but only different grid sizes, Δx=1 and Δx=0.2, respectively. In both figures, one can easily note that in case that using only particle per grid gives poor results, with the large differences from the analytical solutions. However, increasing the number of particles per grid does not seem to necessarily offer improved results greatly as compared to the case two particles per grid is used. Also, as already mentioned, smaller grid size gives good results again.

3.3. Numerical Illustration

To illustrate the model proposed in the study with an application of the one-dimensional transport of radionuclide decay chains, it is assumed that the decay chain begins at the inlet boundary x=0. The decay chain considered in the study is $^{245}Cm \rightarrow {}^{241}Am \rightarrow {}^{237}Np \rightarrow {}^{233}U \rightarrow {}^{229}Th$. The reason why this specific chain is chosen is to show the capability of the model in computing a chain more than three members. All parameters remain the same as in the case of sensitivity study (see Table 1) except the chain decay parameter.

The initial inlet boundary concentrations are unity for parent nuclide and zero for all daughters with Bateman type inlet boundary condition as expressed in Eq.(2c). Figs. 5-7 show the concentration profiles normalized to initial parent concentration as a function of distance at the time of 100 years with different Δx and Δt values with one another . By comparing all these profiles with analytical solutions one can see that all profiles practically coincide with each other. Meanwhile, a more figure for the case Δx=1 and Δt=10 at the time of 1000 years are shown in Fig. 8. For the practical purpose a little larger time steps and grid sizes may be needed. In this aspect the grid size of Δx=1 can be considered to be affordable for the case of very long time span with the time step size of Δt=1 and even Δt=10 as shown both in Fig.

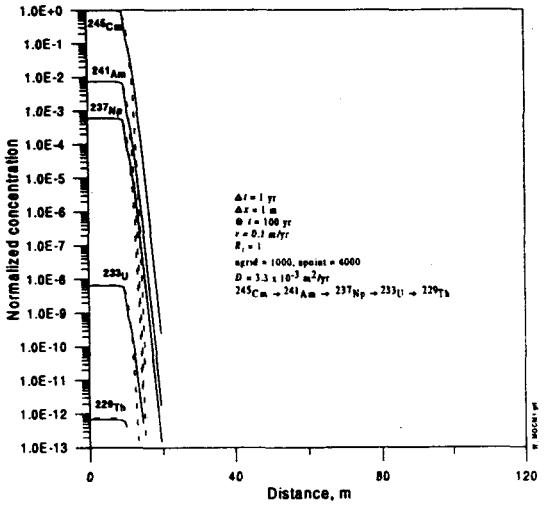

Fig. 5. Profiles at t=100yr for Δx=1 and Δt=1 yr.

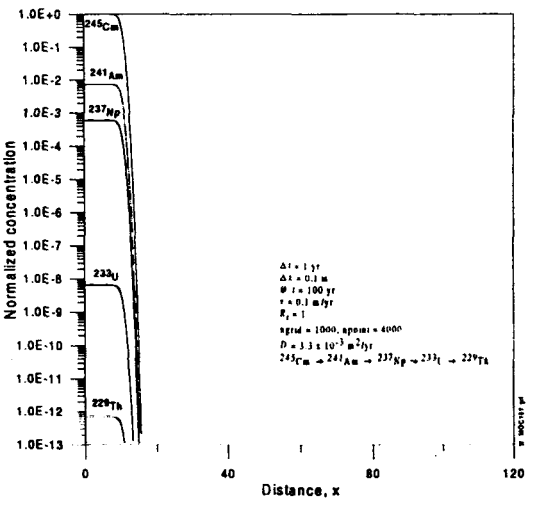

Fig. 7. Profiles at t=1000yrs for Δx=0.1m and Δt=1yr.

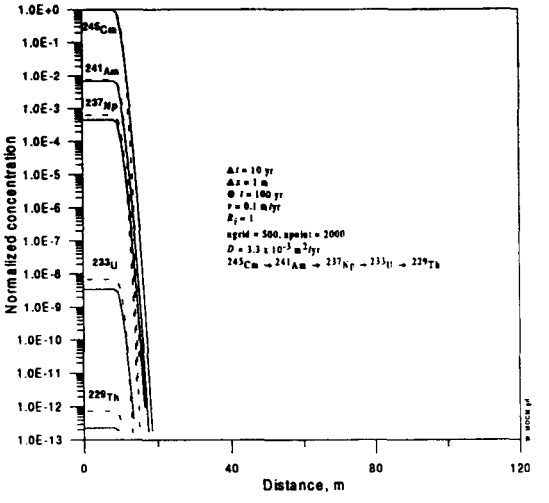

Fig. 6. Profiles at t=100yr for Δx=1 and Δt=10 yrs.

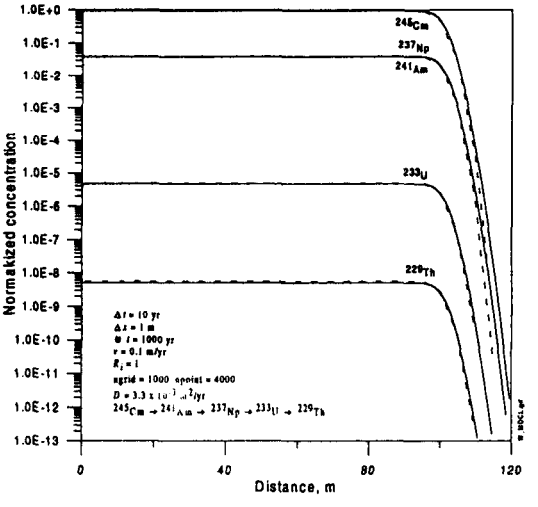

Fig. 8. Profiles at t=1000yr for Δx=1m and Δt=10 yrs.

6 and Fig. 8. A sample calculated breakthroughs as a function of time for illustrative purpose is shown in Fig. 9, which uses $\Delta x=1$ and $\Delta t=10$, respectively, at the distance of 25 m, and the result shows good agreement with analytical solutions.

4. Summary

Through the study the method of characteristics for modeling of nuclide transport has been applied to one-dimensional problem in porous medium. As widely known one of the most distinctive advantages is that the MOC minimizes the numerical dispersion which is distinguished in such numerical scheme as finite difference method and finite element method. Also for the case that decay chain of arbitrary length is involved during transport, MOC is proven to be very effective to solve as shown through several illus-

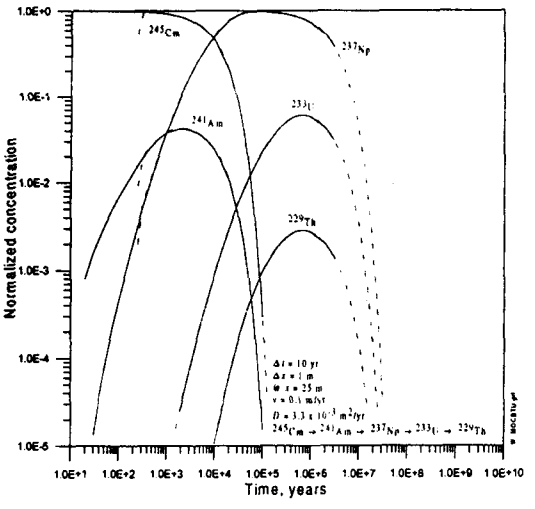

Fig. 9. Breakthroughs at x=25m for Δx=1m and Δt=10 yrs.

Table 1. Parameters Used for Illustration

Nuclide	^{245}Cm	^{241}Am	^{237}Np	^{233}U	^{229}Th
R_i	1	1	1	1	1
$T_{0.5}$(yr)	8500	430	2.14×10^6	1.59×10^5	7430
N_0	1	0	0	0	0

trations.

To solve the advection-dispersion equation with chain decay, the effect of advection is approximated by the particle tracking, which eventually represents the only characteristic of original governing equation. While the advection is simulated by such Lagrangian method based on moving coordinates, the dispersion and decay are solved by standard finite difference scheme which is an Eulerian approach based on the fixed grid at the time point where the advection has just finished. The chain decay is finally implemented by utilizing a superposition concept on account of the linearity of chain decay terms.

Also through the several simulation results compared with available analytical solution, the MOC is possible to be extended to the improved transport problems having both more higher dimensional medium and inhomogeneous media such as fractured rocks.

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