

Three-Dimensional Nuclide Decay Chain Transport Behavior in the Deep Geological Disposal System

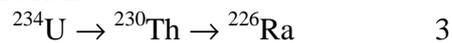
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B3Rch



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Abstract

Using a three-dimensional numerical code, **B3Rch** developed for nuclide transport of an arbitrary length of decay chain in the media including buffer between canister and adjacent rock in a deep geological high-level radioactive waste repository, some illustrative calculations for a case of decay chain of ${}^{234}\text{U} \rightarrow {}^{230}\text{Th} \rightarrow {}^{226}\text{Ra}$, which is one of the most important chain as far as the human environment is concerned, are introduced. A finite difference method utilizing the control-volume scheme is adopted assuming a linear sorption isotherm, nuclide transport due to diffusion in the buffer and the rock matrix, and advection and dispersion along thin rigid parallel fractures existing in a saturated porous rock matrix as well as diffusion through the fracture wall into the matrix. To show how visualization of nuclide behavior can be made to see the effects of buffer and rock matrix on nuclide transport in HLW repository and also to demonstrate usefulness of B3R, several cases of three-dimensional concentration isopleths associated with these disposal system barriers are plotted.

Introduction

The potential repository for the final disposal of high-level radioactive waste (HLW) in Korea is likely to be the one similar to Swedish KBS-3 (1983) located in deep geological crystalline rock formation. According to this concept, the disposal of spent fuel assemblies in canisters are considered to be emplaced in vertical deposition holes. The buffer material is designed to be of low permeability to delay the contact of the waste by groundwater as well as to retard nuclides transporting to the host rocks.

As such HLW repository is to be located in deep geological formations, behavior of chain decaying nuclide in geological media has been an important topic in assessing its performance, recently a series of studies associated with chain decay transport have been carried out numerically by authors (Lee et al., 1989; Lee et al., 1993; Lee et al., 1995; Lee and Lee, 1995; Lee et al., 1996; Lee and Kang, 1997; Lee et al., 1997).

Two- and three-dimensional finite-difference numerical solutions for nuclide transport of an 'arbitrary decay chain length' (i.e., multi-member chain decay) through a buffer and adjacent fractured porous medium by utilizing a control volume method has been developed and the exactness of this solution by comparisons with available analytical solutions has been investigated in a series of works by Lee and Kang (1999a; 1999b) and Lee et al. (1999; 2002).

The purpose of this paper is to illustrate how visualization of nuclide behavior in the host rock can be made for the nuclide transport across the buffer-rock matrix and buffer-fracture interfaces around a canister since the primary application of the model and the B3Rch code is to evaluate and to visualize the effect of nuclide behavior in the HLW repository.

The examples presented in this paper, which are concerned with nuclide transport having decay chains and its effect in the presence of such important mechanisms associated with transport as retardation and rock matrix diffusion, considering in-growth due to daughter nuclides' decay are considered to be sufficient to show usability of B3Rch as an illustrative tool even though rather in-depth sensitivity studies are not made for illustrative purpose.

The Model

After nuclides are escaped from a canister, they will transport by diffusion through the buffer around the canister and eventually will be transferred to the host rock. Fractures in the host rock around the buffer may intersect disposal holes for canister providing groundwater pathways for the hydrogeologic nuclide transport due to advection, whereas porous rock matrix interfaced with buffer will provide diffusive transport pathways for nuclides (see Fig. 1). Since fractures having permeabilities several orders of magnitude higher than the rock matrix itself provide a main hydrogeologic pathway for the transport of the nuclide into the far-field region, the assumption that the rock matrix surrounding the buffer is impervious to nuclide transport has commonly been made. However, recently, studies show that the nuclides are available to diffuse freely across the buffer-matrix boundary and the matrix can play an important role to retard the nuclides requiring rather in depth studies involved.

Thin rigid parallel fractures are embedded in a saturated porous rock matrix by approximating the fractured porous medium as three-dimensional parallel fracture-embedded rock block. The buffer is modeled as a common three-dimensional porous medium. Assuming a linear sorption isotherm, transport for the nuclide l in a saturated fracture can be described by

$$R_l \frac{\partial c_l}{\partial t} + \lambda_l c_l R_l = \frac{\partial}{\partial x} \left(D_L \frac{\partial c_l}{\partial x} - v c_l \right) + \frac{\partial}{\partial y} \left(D_{ly} \frac{\partial c_l}{\partial y} \right) + \frac{\partial}{\partial z} \left(D_{lz} \frac{\partial c_l}{\partial z} \right) + \lambda_{l-1} c_{l-1} R_{l-1}, \quad (1)$$

$$x_b \cos \phi \leq x \leq x_L \quad (0 \leq \phi \leq \pi/2), \quad 0 \leq y \leq b, \quad \begin{cases} x_b \sin \phi \leq z \leq x_b \cap 0 \leq x < x_b, \\ 0 \leq z \leq x_b \cap x \geq x_b \end{cases}, \quad t > 0.$$

Also, unlike fracture, assuming that no advective transport takes place in the buffer as well as in the matrix, the governing equation for nuclide transport in such porous media is

$$R_l^{b,p} \frac{\partial c_l}{\partial t} + \lambda_l c_l R_l^{b,p} = \frac{\partial}{\partial x} \left(\theta_{b,p} D_x^{b,p} \frac{\partial c_l}{\partial x} \right) + \frac{\partial}{\partial y} \left(\theta_{b,p} D_y^{b,p} \frac{\partial c_l}{\partial y} \right) + \frac{\partial}{\partial z} \left(\theta_{b,p} D_z^{b,p} \frac{\partial c_l}{\partial z} \right) + \lambda_{l-1} c_{l-1} R_{l-1}^{b,p},$$

$$\begin{cases} x_c \cos \phi < x < x_c \cos \phi + (x_b - x_c), \quad 0 < y < y_L, \quad x_c \sin \phi < z < x_b \cos \omega, \quad t > 0; & \text{for buffer} \\ x_b \cos \phi < x < x_L, \quad b < y < y_L, \quad x_b \sin \phi < z < x_b, \quad t > 0; & \text{for matrix.} \end{cases} \quad (2)$$

where the boundary conditions are zero concentration gradient (Neuman-type) boundary conditions as represented in Eq. (3) at all boundaries only except at the canister surface interface, *i.e.*, except for the inlet:

$$\frac{\partial c_l}{\partial x} = 0, \quad x = x_L \quad (3)$$

For initial and boundary conditions associated with the inlet of the buffer, nuclide decay and transformation of the parent nuclide into its daughter products are considered together by Bateman's decaying source. Therefore the concentration at the inlet boundary for the l -th component of the decay chain, $\tilde{c}_l(t)$, which is written as

$$\tilde{c}_l(t) = \sum_{m=1}^l B_{lm} e^{-\lambda_m t} \quad (4a)$$

where

$$B_{lm} = \sum_{n=1}^m c_n^0 \prod_{k=n}^{l-1} \lambda_k \left\{ \prod_{\substack{j=n \\ j \neq m}}^l (\lambda_j - \lambda_m) \right\}^{-1} \quad (4b)$$

and c_l^0 denotes the initial concentration due to inventory at the inlet.

The full description and notations for the finite discretization procedure adopting control volume approach is represented in detail in the previous paper (Lee et al., 2002).

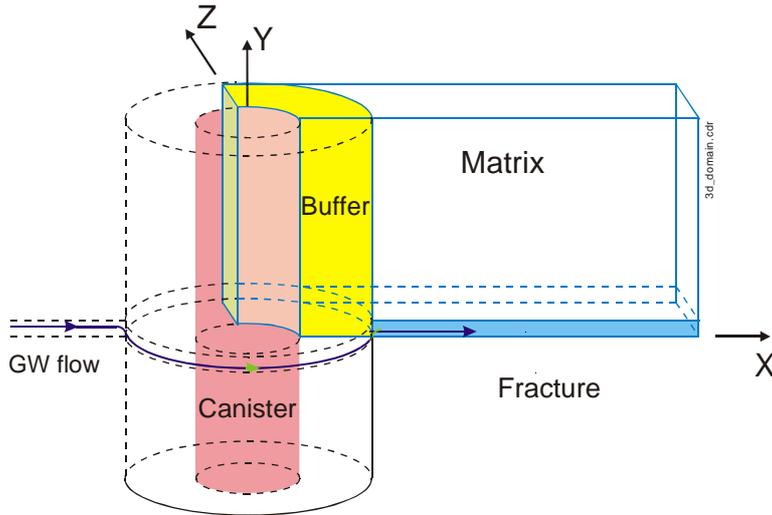


Fig. 1. Schematic view of a deposition hole intersected by a fracture in a potential HLW repository tunnel

Illustration

The volume- or face plots of concentrations of nuclides ($^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$) in the media around the quartile section of a canister (see Fig. 1), normalized to its initial concentration at the inlet boundary surface of the canister where initial concentrations are to be decayed time-dependently according to Bateman equation (Eqs. (4) and (6) in Table 4), as a function of distances in the x -, y - and z -directions at time equal to 8×10^3 years are shown in Figs. 2 through 9. Even though only for three steps of decay chain of $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$ are limited to current study, B3Rch is not limited by the specific number of members in the chain.

Figs. 2 through 3, among others, considered as base case scenario for each nuclide, show the isopleths in case there are matrix diffusions existing from the fracture, but without any retardation through whole media. On the other hand, Figs. 4 through 6 show each case respectively that retardations for all media are considered, whereas matrix diffusion is still associated, though. The other plots in Figs. 7 through 9 are ones for the case that matrix diffusion is not involved.

By investigating, one can easily find that there are prominent effects of retardation and matrix diffusion phenomena on the concentration profiles compared to base case plots: Plots for nuclide concentrations for the case retardations are considered for all media (Figs. 4 to 6) show rather slowly spreading isopleths, compared to the baseline case with no retardation (Figs. 2 to 3). For the case there is no transverse matrix diffusion through the fracture wall, Figs. 4 to 6 show their remarkably dominant concentration isopleths only along the fracture (around $y = 0$) very naturally since after they leave buffer region nuclides in the fracture travel relatively faster than those in the rock matrix.

The nuclides entering the fracture are taken away by advection and dispersion due to groundwater flow that takes place only in the fracture whether there is no loss or loss of nuclide concentration due to diffusion into the rock matrix from the fracture.

For all figures depicted through the study, nuclides are considered to be supplied both across buffer-matrix interface as well as through the inlet of the fracture.

In summary, with the advection-dispersion parameters chosen, these calculations visualizes the isopleths that ^{234}U travels physically well around the canister, buffer and other

surrounding media.

Table 1. Spatial increments for the control volume

Δx_i , cm $i=1, 2, 3, \dots, 100$	7($i=1 \dots 5$), 10($i=6 \dots 10$), 15($i=11 \dots 50$), 20($i=51 \dots 100$)
Δy_j , cm $j=1, 2, 3, \dots, 20$	0.006(= $b/2$), 0.009, 0.015, 0.025, 0.045, 0.07, 0.13, 0.25, 0.45, 1($j=10 \dots 20$)
Δz_k , cm $j=1, 2, 3, \dots, 10$	7($k=1 \dots 5$), 10($k=6 \dots 10$)

As seen in the Table 1, the number of control volumes used is 100 along the fracture axis in the x -direction, 20 in the y -direction into the matrix, and 10 in the horizontal z -direction, giving a total of $100 \times 20 \times 10$ control volumes. Parameters used are also listed in Tables 2 and 3.

Table 2. Nuclide data

Nuclide	$t_{0.5}$, yr	R_l, R_l^b, R_l^p	c_l^0
^{234}U	2.47×10^5	0. (120.)	$c_0 = 1.0$
^{230}Th	$8. \times 10^4$	0. (120.)	$c_0 = 0.0$
^{226}Ra	1600.	0. (120.)	$c_0 = 0.0$

Table 3. Parameters used.

Parameter	Value
$2b$	120 μm
θ_b	0.1
θ_f	1.0
θ_p	0.4
α_L	0.76 m
v	0.75 m/yr
$(\theta D_x^p) = (\theta D_y^p) = D^*$	$8.64 \times 10^{-6} \text{ m}^2/\text{yr}$
D_L	$\alpha_L \cdot v + D^*$
buffer thickness ($= x_b - x_c$)	35cm

Table 4. Initial and boundary conditions.

I.C.	$c_l(x, y, z; t = 0) = 0, \quad l = 1, 2, 3$ (5)
B.C. (@ inlet)	$c_l(x = x_c \cos \phi, 0 \leq y \leq y_L, 0 \leq z \leq x_b; t) = \tilde{c}_l,$ $l = 1, 2, 3, \dots$ (6a)
	$v c_l(x = x_c \cos \phi, 0 \leq y \leq y_L, 0 \leq z \leq x_b; t) - D_L \frac{\partial c_l}{\partial y} = v \tilde{c}_l,$ $l = 1, 2, 3, \dots$ (6b)

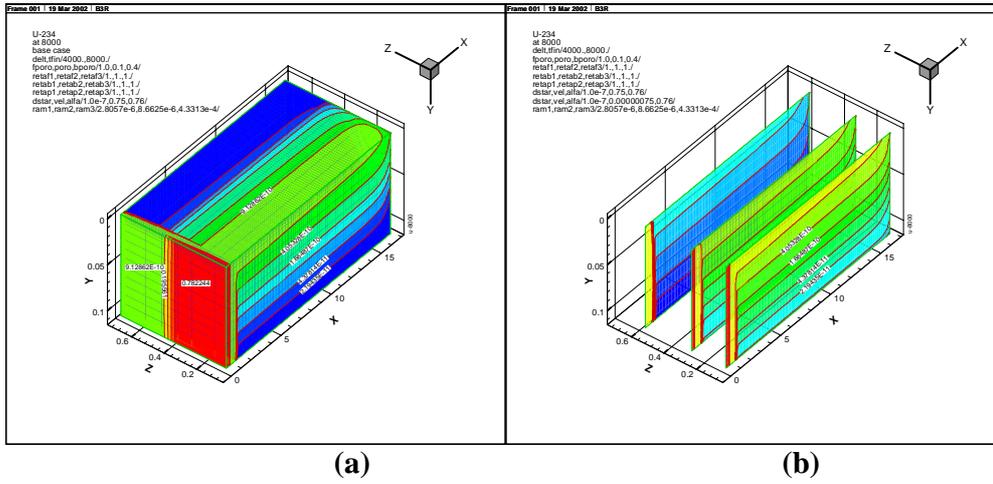


Fig. 2. Volume plot (a) and face plot (b) for ^{234}U at time of 8×10^3 years

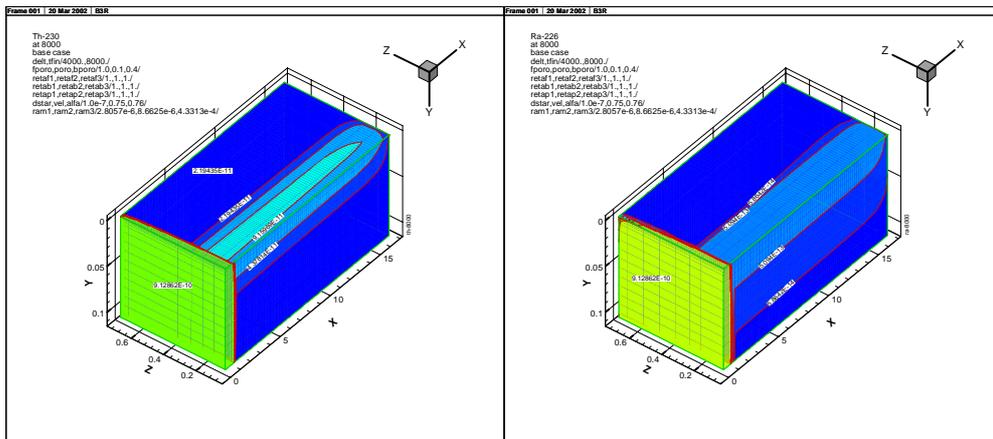


Fig. 3. Volume plot (a) and face plot (b) for ^{230}Th at time of 8×10^3 years

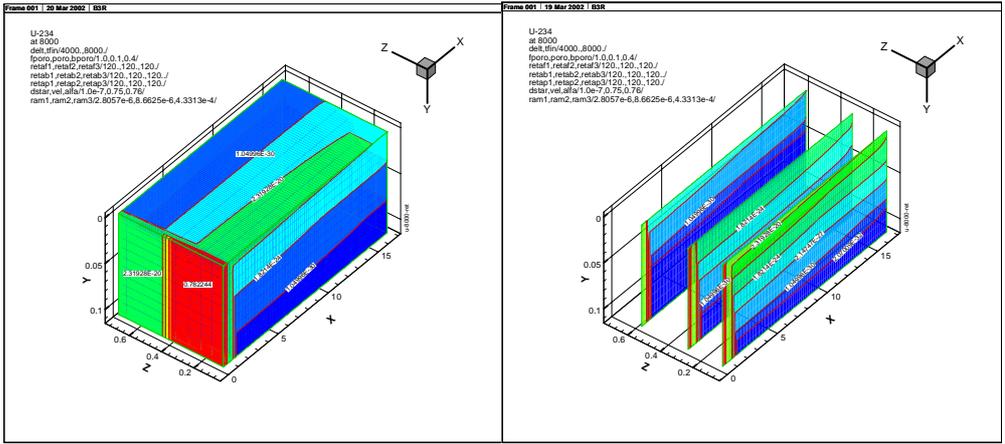


Fig. 4. Isopleths of ^{234}U at time of 8×10^3 years: retardation considered

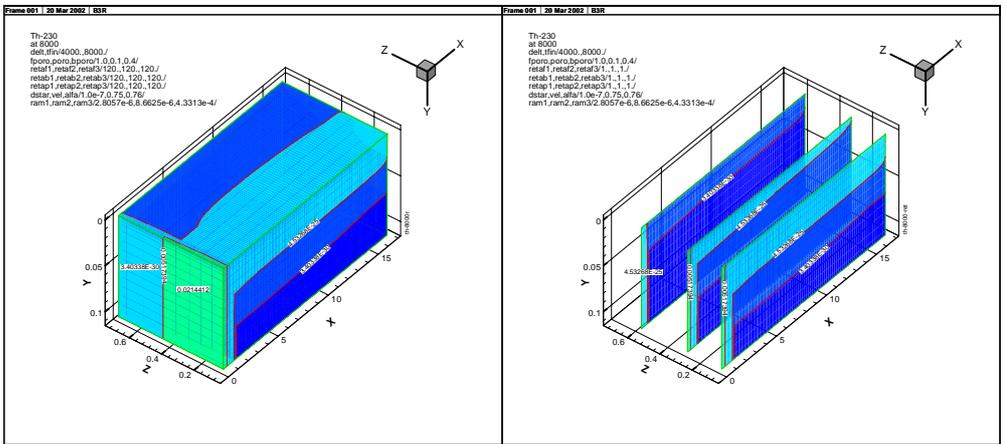


Fig. 5. Isopleths of ^{230}Th at time of 8×10^3 years: retardation considered

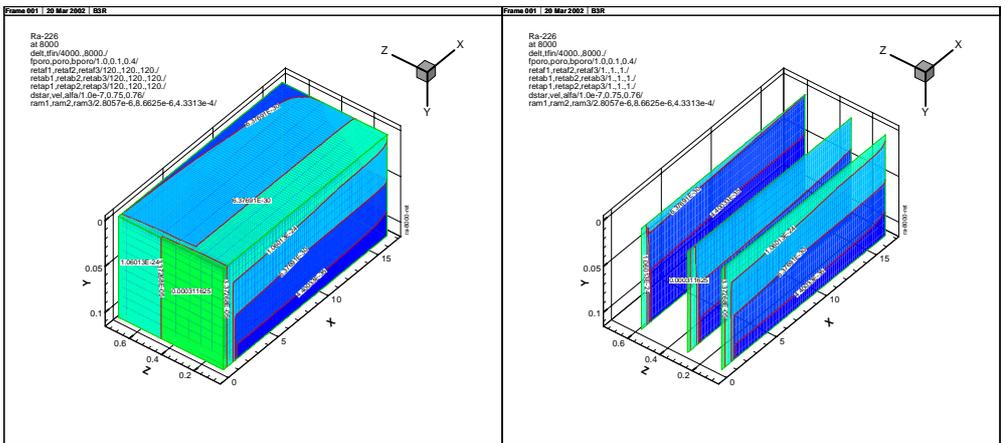


Fig. 6. Isopleths of ^{226}Ra at time of 8×10^3 years: retardation considered

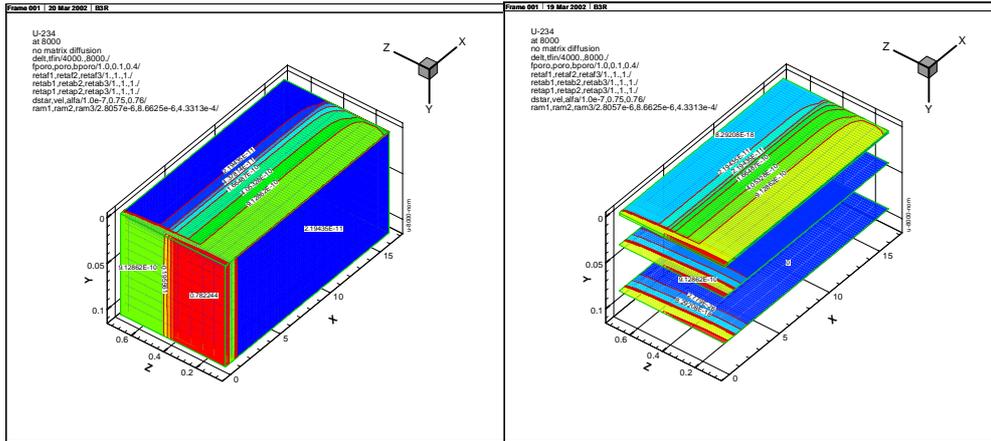


Fig. 7. Isopleths of ^{234}U at time of 8×10^3 years: no matrix diffusion available

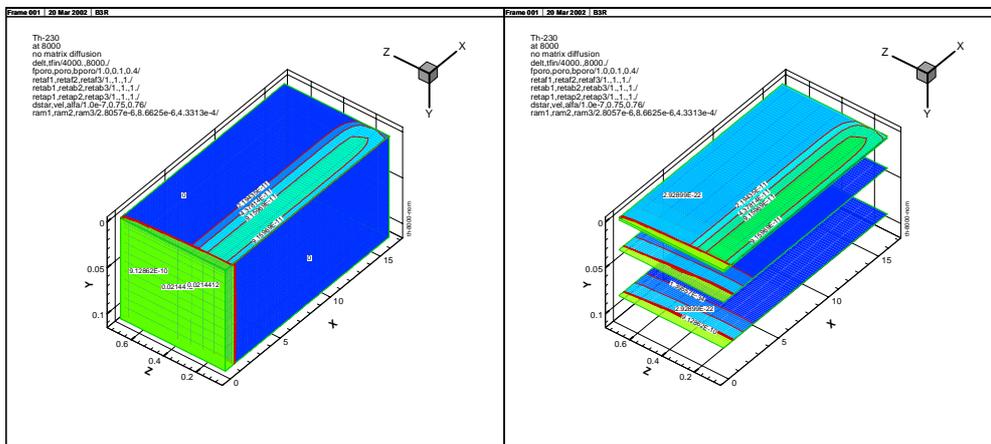


Fig. 8. Isopleths of ^{230}Th at time of 8×10^3 years: no matrix diffusion available

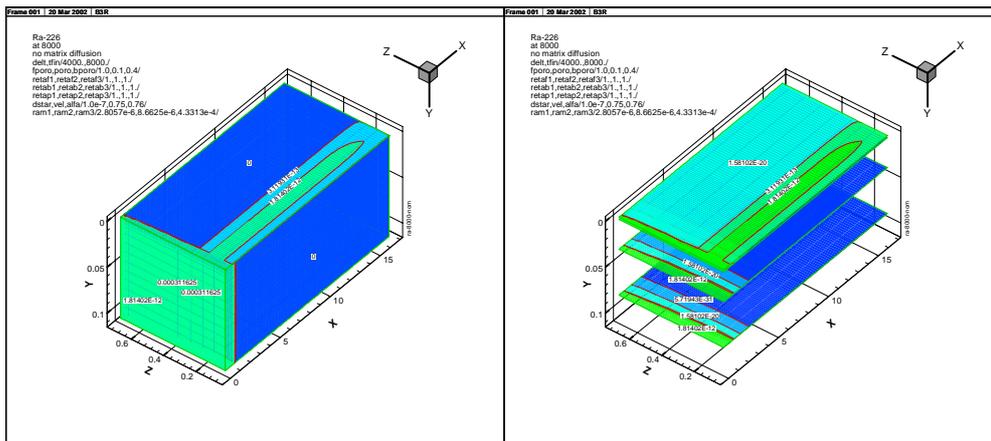


Fig. 9. Isopleths of ^{226}Ra at time of 8×10^3 years: no matrix diffusion available

Concluding Remarks

A three-dimensional behaviors of decay chain of $^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$, which is one of the most important chain in view of safety of human environment are introduced utilizing a control volume method and a computer code, B3Rch have been introduced to illustrate multi-member decay chain transport in HLW repository system.

Such transport behavior of nuclide having decay chains and its effect in the presence of buffer and rock matrix diffusion, considering in-growth due to daughter nuclides' decay is an important topic to show the importance of barrier function in view of HLW repository safety. To show three-dimensional visual transport behavior, some volume and surface plots for nuclide concentration isopleths were shown.

The model, which is an extension of previous work by Lee and Kang (1999a; 1999b) who developed the two-dimensional model for decay chain transport in a composite media of the buffer and the fractured porous medium is based on a physically exact formulation utilizing a control volume method and then the differential governing equation is directly integrated over each control volume. This kind of work believes to be very useful especially when a visualization is needed in order to represent the nuclide behavior around the canister and there needs a graphical representation of the nuclide behavior around the deep geological repository to investigate transport phenomena through the geologic media involved in the safety assessment of such repository and case studies to see what happens when varying properties of such barriers as the buffer and the rock matrix are applied to as well.

Using various computational results from B3Rch, real transport phenomena in a fracture are expected to be visualized not only for scientific communities but also for general public who still have negative feelings on the safety of a radioactive waste repository.

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