

RADIONUCLIDE TRANSPORT IN GEOLOGICAL ENVIRONMENT MODELED IN THE GERA CODE WITH AN ACCOUNT TAKEN OF RADIOACTIVE DECAY CHAINS: NUMERICAL SCHEME VERIFICATION AND CALCULATION SPECIFICS

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The paper describes a numerical scheme implemented in the GeRa code to solve the problem of radionuclide transport in porous medium taking into account radioactive decay chains. It is based on a scheme of splitting by physical processes and the use of separate modules for solving transport and radioactive decay chain problems. Verification of the numerical scheme was performed using an analytical solution for a specific case of the problem formulation in a homogeneous porous medium. The study explores the error introduced into the numerical solution by the use of the splitting scheme. The study provides recommendations regarding the selection of appropriate parameters for the numerical scheme to calculate the transport of nuclide chains with different sorption properties.

Keywords: *radionuclide transport in geological environment, radioactive decay, GeRa code, radioactive waste.*

Introduction

Radionuclide groundwater transport calculations under the long-term safety assessment of radioactive waste disposal facilities (RWDF) shall account for the radioactive decay with chains of daughter radionuclides formed along the way [1]. The GeRa code [2] designed to solve three-dimensional flow and mass transport problems evaluate the radioactive decay chains within a specialized module [3]. Thus, a splitting scheme by physical processes is used to introduce the decay chains into transport models: first, the transport problem is solved based on various numerical schemes of the finite volume method, then, based on the obtained concentrations, the radioactive decay is calculated in each cell of the computational grid. It should be noted that the splitting scheme inevitably introduces an error being directly proportional to the time step. Thus, the radionuclides may reach the points remote from the contamination source overly early or

the peak concentrations at these points may turn to be underestimated.

This study verifies the developed model, evaluates its features related to numerical implementation and provides recommendations for its practical application in the safety assessment of waste repositories and other nuclear facilities. Therefore, in keeping with the algorithm discussed in [4], it presents an analytical solution in its explicit form and then compares it with the numerical solution in the GeRa code; it explores the calculation features specific for transport problems with radioactive decay given considerable difference in the sorption capacity of radionuclides included in the chains; it provides recommendations on the selection of the calculation time steps, discusses the numerical error introduced by the splitting scheme and provides methodological recommendations on error reduction.

Mathematical model and calculation scheme

The equation of advective-diffusion transport in a porous medium for a parent nuclide can be presented as follows (see, for example, [5]):

$$\theta R_1 \frac{\partial C_1}{\partial t} - \nabla(D\nabla C_1) + \nabla(\bar{u}C_1) + \theta R_1 \lambda_1 C_1 = 0. \quad (1)$$

where C_1 is the specific activity of the parent nuclide, θ is the porosity of the medium, D is the diffusion-dispersion tensor, \bar{u} is the filtration (Darcy) velocity, λ_1 is the decay constant, R_1 is the sorption retention coefficient for the radionuclide that can be calculated as follows:

$$R_1 = 1 + \frac{k_{d1}\rho_b}{\theta}, \quad (2)$$

where k_{d1} is the sorption distribution coefficient of the parent nuclide, ρ_b is the bulk density of the porous matrix.

The equation for the transport of the i -th daughter radionuclide can be written using similar quantities:

$$\theta R_i \frac{\partial C_i}{\partial t} - \nabla(D\nabla C_i) + \nabla(\bar{u}C_i) + \theta R_i \lambda_i C_i - \theta \sum_{j<i} R_j p_{ji} \lambda_j C_j = 0. \quad (3)$$

where p_{ji} is the probability that the decay of the j -th nuclide produces the i -th nuclide (in case of chain branching).

In the GeRa code, radionuclide transport and the decay along the chains is calculated in two interconnected modules. The first one applies various finite volume method schemes [6] to solve the problem numerically, the second one is based on the analytical solution of the radioactive decay equation:

$$N_n = N_{10} \prod_{k=1}^{n-1} (\lambda_k p_k) \left(\sum_{i=1}^n \frac{\exp(-\lambda_i t)}{\prod_{\substack{j=1 \\ j \neq i}}^n (\lambda_j - \lambda_i)} \right), \quad (4)$$

where N_{10} is the initial number of parent isotope nuclei. It should be noted that N is introduced both as the dissolved substance and as the substance sorbed on the rock. Accordingly, when the decay step is calculated, the equilibrium between the solution and the rock is recalculated. Formula (4) is valid assuming that only the parent nuclide atoms, from which the chain starts, exists at the initial time point. In the case where a general solution is required for non-zero initial quantities N_{n0} , where $n > 1$, it can be calculated by adding similar expressions to this formula for shorter chains starting with the corresponding N_{n0} . Information on the composition of the chain and the decay constants is loaded into GeRa from the nuclear data library [7].

Radionuclide concentrations are recalculated at each time step in each cell of the computational grid. These can be calculated both as radionuclide concentrations and the specific activities.

Formulas providing analytical solution and verification of the numerical scheme

To verify the correctness of a code, one may, for example, calculate a problem having an analytical solution. The study [4] presents an algorithm providing an analytical solution to equations (1)–(3) supplemented by a certain formulation of boundary and initial conditions based on the Laplace transform. The porous medium is assumed homogeneous. Relative to hydrodispersion, the diffusion contribution to the transport is neglected. According to this algorithm, one may put down the explicit solution for a one-dimensional transport problem involving two adjacent radionuclides from a single radioactive decay chain in a semi-infinite column along the x axis. The filtration velocity accounts for $u > 0$. Fixed values of the radionuclide concentrations are specified at the boundary of the column:

$$C_1(0, t) = C_1; \quad (5)$$

$$C_2(0, t) = C_2.$$

If the diffusion is neglected, the diffusion-hydrodispersion tensor D can be presented as follows:

$$D = \alpha u, \quad (6)$$

where α is the dispersivity. At the initial time point, concentrations of radionuclide transferred in the column volume are zero.

The analytical solution to this problem can be written as follows:

$$C_1(x, t) = C_1 A_1(x, t),$$

$$C_2(x, t) = C_2 A_2(x, t) + \frac{R_1 \lambda_1 C_1}{R_2 \lambda_2 - R_1 \lambda_1} (A_1(x, t) - A_2(x, t) - B_1(a, x, t) + B_2(a, x, t)). \quad (7)$$

The following auxiliary functions were introduced here:

$$A_i(x, t) = \frac{1}{2} \left(e^{\frac{x(1-v)}{2\alpha_x}} \operatorname{erfc} \left(\frac{R_i x - uv t}{\sqrt{4\alpha_x u R_i t}} \right) + e^{\frac{x(1+v)}{2\alpha_x}} \operatorname{erfc} \left(\frac{R_i x + uv t}{\sqrt{4\alpha_x u R_i t}} \right) \right);$$

$$v = \sqrt{1 + \frac{4\alpha_x R_i \lambda_i}{u}};$$

$$B_i(x, t) = \frac{e^{-at}}{2} \left(e^{\frac{x(1-\mu)}{2\alpha_x}} \operatorname{erfc} \left(\frac{R_i x - u\mu t}{\sqrt{4R_i \alpha_x u t}} \right) + e^{\frac{x(1+\mu)}{2\alpha_x}} \operatorname{erfc} \left(\frac{R_i x + u\mu t}{\sqrt{4R_i \alpha_x u t}} \right) \right);$$

$$\mu = \sqrt{1 + \frac{4\alpha_x R_i (\lambda_i - a)}{u}};$$

$$a = \frac{R_2 \lambda_2 - R_1 \lambda_1}{R_2 - R_1}. \quad (8)$$

To test the numerical scheme in GeRa, the transport in a column filled with clay was calculated for a chain involving two radionuclides $^{234}\text{U} \rightarrow ^{230}\text{Th}$. Table 1 presents the parameters of the problem. The model was implemented in a quasi-one-dimensional formulation: the column was divided into a uniform hexahedral grid along the x -axis.

Table 1. Parameters of the verification test with a nuclide chain transport

Parameter	Value
Column length	10 m
Δx (grid cell size)	0.1 m
Δt (time step)	25 years
θ (porosity)	0.4
α (dispersivity)	1 m
ρ_b (bulk density)	2,000 kg/m ³
u (filtration velocity)	0.04 m/day
k_d (^{234}U)	1.6 m ³ /kg
k_d (^{230}Th)	5.8 m ³ /kg
$T_{1/2}$ (^{234}U)	245,878 years
$T_{1/2}$ (^{230}Th)	75,438 years
C_1 (^{234}U concentration at the left edge of the column)	1 mol/l
C_2 (^{230}Th concentration at the left edge of the column)	0 mol/l

Fig. 1 shows the concentration distribution in the column for both radionuclides at the time point of 1,000 years. The analytical solution (7)–(8) is compared with the calculated result obtained in GeRa

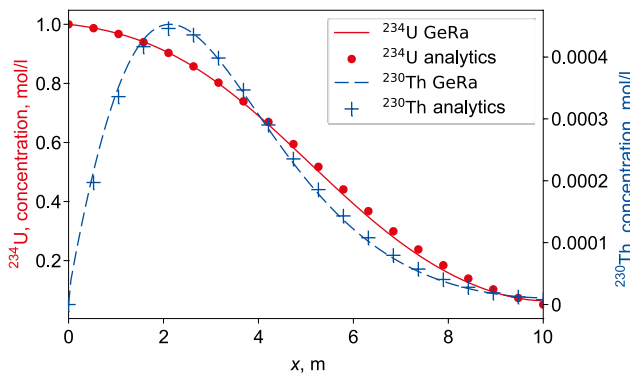


Fig. 1. Distribution of ^{234}U and ^{230}Th concentrations along the column length at the time point of 1,000 years: GeRa calculation results and the analytical solution

using an explicit discretization scheme for the advection operator of the second-order accuracy in space. Good agreement between the results appears to be quite evident.

Coupling error assessment

The transport and decay modules coupling in the GeRa code is based on a splitting scheme [8]: at each time step, a transport step and a calculation step for the decay along the chain are performed sequentially. [3] evaluates the numerical error introduced by such coupling when calculating the chain transport for two nuclides. Its value for the daughter nuclide can be calculated as follows:

$$err = \theta \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(e^{-\lambda_1 \Delta t} - e^{-\lambda_2 \Delta t} \right) \frac{\partial C_1^s}{\partial t} \left(R_1 - \frac{R_1^2}{R_2} \right), \quad (9)$$

where C_1^s is the concentration of the first nuclide reached when the transport step in the splitting scheme is done.

From expression (9) it follows that the error is not zero when the sorption retardation coefficients for the parent and daughter nuclides are different. As the time step tends to zero, the following expression can be obtained:

$$\lim_{\Delta t \rightarrow 0} err = \theta \lambda_1 \frac{\partial C_1^s}{\partial t} \left(R_1 - \frac{R_1^2}{R_2} \right) \Delta t. \quad (10)$$

The error has the first order of approximation in time, which is common for this type of physical processes and the corresponding splitting schemes [8]. This is acceptable in our case, since the discretization of the transport equation in time in the GeRa code also has the first order of approximation.

Based on expressions (9)–(10), it is clear that considering the calculation error, the proximity in the chain of a relatively long-lived strongly sorbed nuclide to a short-lived weakly sorbed one should be treated as the most “dangerous” case. Considered below as an example is the transport of the following chain observed in a column filled with sand: $^{243}\text{Am} \rightarrow ^{239}\text{Np} \rightarrow ^{239}\text{Pu}$. At the initial time point, the radionuclide ^{243}Am , whose activity amounts to 10^{10} Bq, enters the first cell of the quasi-one-dimensional column grid. Table 2 presents the parameters of the problem.

Considered below are various combinations of numerical schemes and time steps. The calculation results in the dependence between the specific activities of nuclides in solution and time at point $x = 50$ m.

Fig. 2 shows the final indicators for the explicit high-order scheme with a time step $\Delta t = 5$ days. The step was selected so that the Courant criterion could be met for the fastest radionuclide in the

Table 2. Model parameters

Parameter	Value
Column length	50 m
Δx (grid cell size)	1 m
θ (porosity)	0.43
ρ_b (bulk density)	1,600 kg/m ³
u (filtration velocity)	1 m/day
k_d (²⁴³ Am)	1.9 m ³ /kg
k_d (²³⁹ Np)	0.005 m ³ /kg
k_d (²³⁹ Pu)	0.55 m ³ /kg
$T_{1/2}$ (²⁴³ Am)	2.69·10 ⁶ days
$T_{1/2}$ (²³⁹ Np)	2.36 days
$T_{1/2}$ (²³⁹ Pu)	8.81·10 ⁶ days

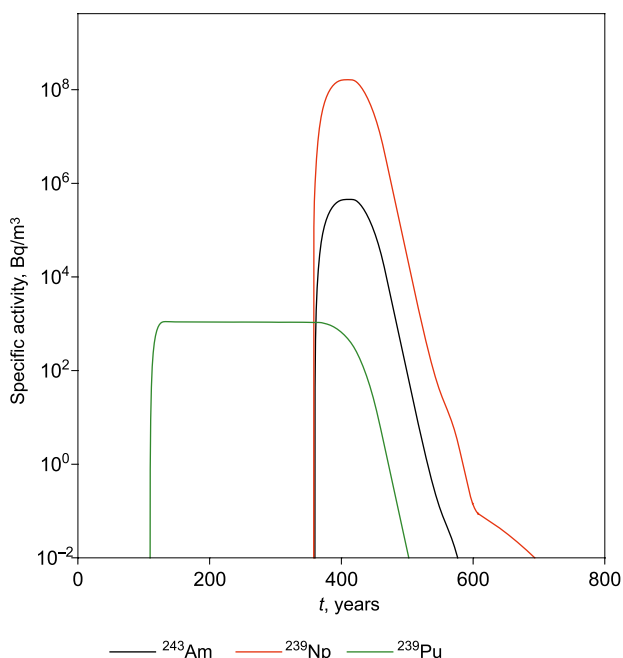


Fig. 2. Dependence between the specific activity of radionuclides at the observation point and time (high-order explicit scheme, $\Delta t=5$ days)

chain — ²³⁹Np, which physically means that within a single time step, a particle of the matter should not move more than over a single spatial step [9]. In case of explicit schemes, this condition is required to be met to provide stable operation.

It is evident that, despite the high transport rate, ²³⁹Np, due to its short half-life, tends to be in equilibrium with its parent ²⁴³Am. Thus, they reach the observation point simultaneously (the maximum is reached at ~ 400 years). ²³⁹Pu appears there first in ~ 120 years. In the time interval between the first appearances of ²³⁹Pu and ²⁴³Am (in the range of 120–360 years), the specific activity of ²³⁹Pu is practically constant since plutonium, which appeared later,

“tears away” from the americium and neptunium activity cloud and arrives at the observation point.

Let us now consider the result obtained based on the implicit first-order scheme in space with the same time step (Fig. 3).

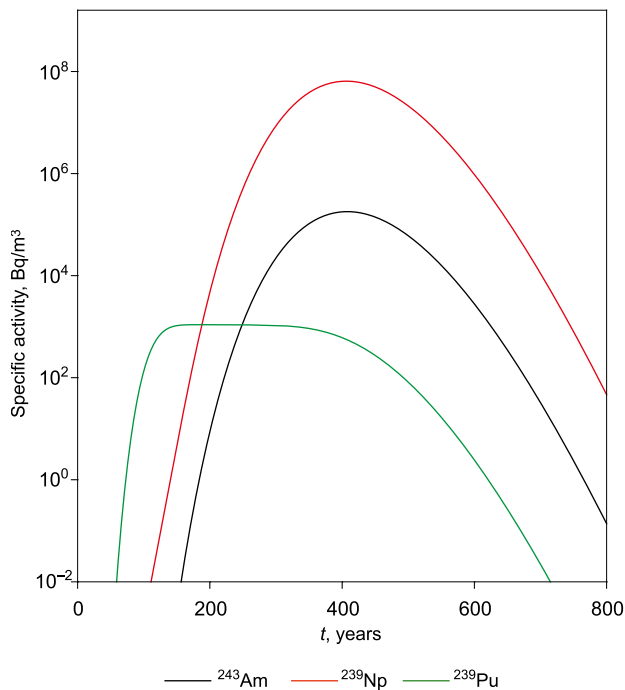


Fig. 3. Dependence between the specific activity of radionuclides at the observation point and time (implicit scheme, $\Delta t=5$ days)

It is evident that the peaks are noticeably (the maximum specific activities for neptunium and americium are ~2.5 times smaller) spread out due to diffusion. The time of their arrival remains constant, but the growth of plutonium activity is observed in a ~ 50 years’ time (in the model with an explicit high-order scheme, in ~ 100 years’ time).

Considered below are the calculation results considering the implicit scheme with a time step of $\Delta t=400$ days (Fig. 4), which ensures the fulfillment of the Courant criterion for plutonium according to the time parameter, and since the equilibrium activity of neptunium depends on the one of americium, such a step may seem adequate under this model.

Although the peak times and the specific activity maxima remain the same, noticeable values for plutonium appear at the observation point from the very beginning of the calculation, and only starting from the time point of ~ 60 years (the inflection point is designated on the graph as T1) does this dependence take the shape similar to the one plotted based on previous calculations. This phenomenon is explained by the method applied to couple the decay modules along the chains and the transport.

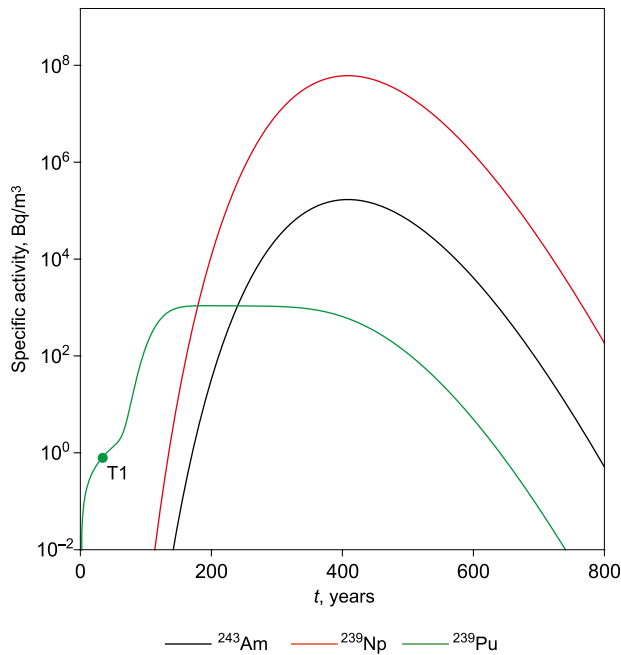


Fig. 4. Dependence between the specific activity of radionuclides at the observation point and time (implicit scheme, $\Delta t = 400$ days)

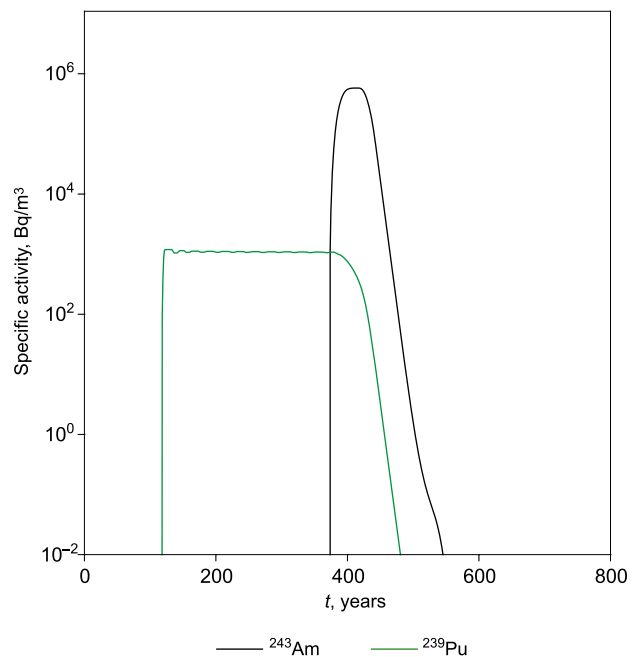


Fig. 5. Dependence between the specific activities of radionuclides at the observation point and the time (high-order explicit scheme, $\Delta t = 400$ days)

Already at the first steps in time, the fast-migrating neptunium manages to run through the entire column and turns into plutonium at the decay stage. Thus, plutonium ends up where it should not be.

Now let's further exclude neptunium from the model (assuming that americium decays immediately and produces plutonium) and use an explicit high-order scheme with a step of $\Delta t = 400$ days in our calculations. It should be noted that it's impossible to perform calculations with such a time step for neptunium since it violates the Courant criterion for this nuclide causing calculation instabilities. The calculation result is shown in Fig. 5.

The result is practically the same as the one obtained for $\Delta t = 5$ days (Fig. 2). Small fluctuations in the specific activities of plutonium are caused by the use of a high-order scheme for the distributed source problem; the introduced error amounts to $\sim 5\%$.

Thus, when calculating the radionuclide geomigration problem with consideration given to radioactive decay chains some inadequate results can be obtained if the time step and the calculation scheme are selected inaccurately. To avoid such errors in these calculations, explicit high-order spatial transport schemes should be used whenever possible. However, this approach imposes some restrictions on the time step, which may considerably increase the calculation time in case of complex models with a large number of cells in the grid. To avoid such a difficulty, weakly sorbed daughter nuclides should be excluded from the calculations whenever possible. This is acceptable if their

half-life is much shorter than the selected time step. Then it can be guaranteed that the considered daughter radionuclide is in equilibrium with the parent nuclide (commonly this is assumed to happen in ~ 10 half-lives [10], accordingly, the exclusion condition can be presented as follows $T_{1/2} < 0.1\Delta t$), and it is sufficient to add the equilibrium activity of the daughter radionuclide to the final result.

Conclusions

The GeRa calculation code implements a numerical modeling algorithm for the radionuclide geomigration with an account taken of the radioactive decay chains. The model was verified on a problem with an analytical solution. The process splitting scheme used in the algorithm introduces certain computational error: the more the sorption properties of the parent and daughter nuclides differ, the greater the error. To minimize it in practical calculations performed in the GeRa code, the following recommendations can be provided:

- an explicit scheme of the second order of accuracy in space is preferable for the discretization of the advection operator;
- the time step should be selected so that the Courant criterion is met for the least sorbed radionuclide;
- it is better to exclude weakly sorbed rapidly decaying radionuclides from the transport calculation and take them into account in the final result in equilibrium with the parent nuclide.

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Bibliographic description

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