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DRBEM simulation of radionuclide transport near nuclear waste repository

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Abstract

Numerical modelling and simulation represent essential tools for evaluation of the safety of nuclear waste repositories. The Dual Reciprocity Boundary Element method (DRBEM) is used in this paper to solve the Darcy–Brinkman equation, and coupled species transport equations of the Pu-241 decay chain near the hypothetic repository. For this purpose, the very recently developed unified DRBEM formulation for nonlinear coupled transport phenomena has been applied. Fully implicit time-discretization, Laplace equation fundamental solution weighting, scaled augmented thin plate spline based dual reciprocity transformation, and constant discontinuous boundary elements are used for space-discretization in two dimensions. The simulations will be used in independent performance assessment of the foreseen Slovenian low and intermediate level radioactive waste repository.

1 Introduction

The radioactive waste management is a very important humanity issue. The design goal is a repository that society will have a high confidence in [1]. The heart of the safety case for a repository lies in computer models that predict the long-term fate of the disposed radionuclides. This type of modelling [2,3] is reffered to as Performance Assessment (PA). This is a very complex task with many technical disciplines involved [4], covering numerous physical, chemical and biological processes such as: (i) Corrosion of waste containers, (ii) Degradation of the waste materials in the presence of groundwater, (iii) Physical and chemical processes that can result in the exposure of radionuclides to the groundwater within the repository, (iv) Transport processes that could result in migration of released radionuclides through the geosphere to the biosphere, (v) Physical, biological, and anthro-

pogenic processes in the biosphere that could affect the exposure, uptake and dose to human or ecosystem receptors, (vi) The effects on all systems due to climate change, human intrusion, or other external factors.

In PA we need to qualitatively understand and quantitatively conservatively estimate the paths and intensity of pollutants transfer from the repository to geosphere and further to biosphere. The source of the pollutant is in the repository, which is made of concrete and it is representing an engineering barrier. In this paper we focus on computation of transport of radioactive pollutants from the repository to near-field host geological system [5]. The schematics of the problem is shown in Figure 1.

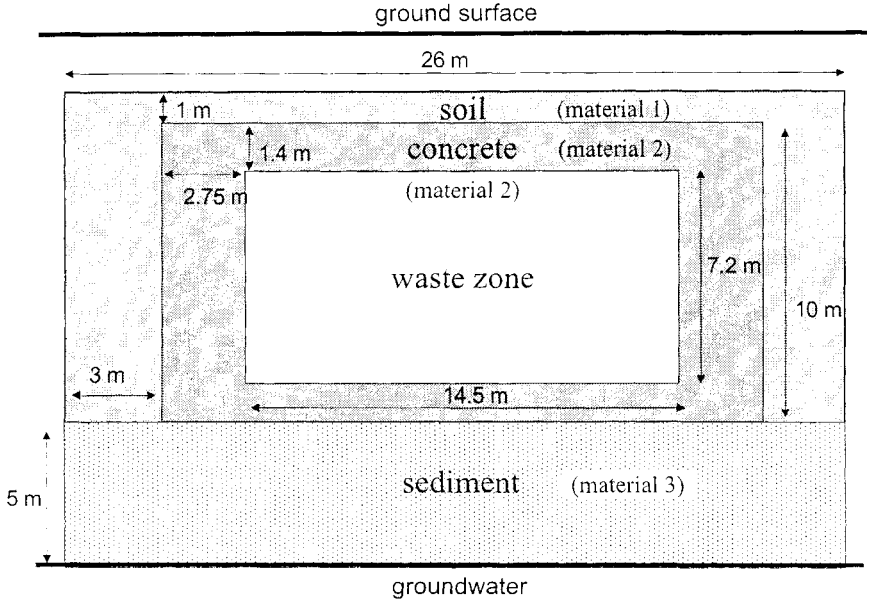

Figure 1: Schematics of the hypothetical repository and the near-field surrounding.

2 Governing equations

Four governing equations are used in this paper in order to demonstrate the complete coping with the basic ideas and the key elements of the solution of the near-field problem by the DRBEM. The equations are posed on a connected fixed domain Ω with boundary Γ occupied by three different materials: the soil, the backfill with waste zone, and the sediment. All three involved materials are modelled as Darcy-Brinkman [6] saturated porous media with the bulk density ϱ_b, porosity ϵ, permeability K, distribution coefficient K_d, longitudinal α_L and transversal α_T dispersivity, and molecular diffusion coefficient D_p. The radionuclide transport is carried out by groundwater with viscosity μ and density ϱ. The groundwater flow is

described by the following two mass and momentum conservation partial differential equations for the seepage velocity $\mathbf{v}$

$$\frac{\partial}{\partial t}\varrho + \nabla \cdot (\varrho\,\mathbf{v}) = 0 \tag{1}$$

$$\frac{\partial}{\partial t}(\varrho\,\mathbf{v}) = -\nabla P - \frac{\mu}{\mathcal{K}}\mathbf{v} + \nabla \cdot (\mu\,\nabla\mathbf{v}) \tag{2}$$

Only two radionuclides are taken into consideration because of the conference paper limitations. Transport of Np-237 is described by

$$\frac{\partial}{\partial t}C_1 + \nabla \cdot (\mathbf{v}\,C_1) = \nabla \cdot (\mathbf{D}_1\,\nabla C_1) - \lambda_1\,R_1\,C_1 + \frac{\partial}{\partial t}\Upsilon_1 \tag{3}$$

and the transport of U-233 is described by

$$\frac{\partial}{\partial t}C_2 + \nabla \cdot (\mathbf{v}\,C_2) = \nabla \cdot (\mathbf{D}_2\,\nabla C_2) - \lambda_2\,R_2\,C_2 + \lambda_1\,R_1\,C_1 + \frac{\partial}{\partial t}\Upsilon_2 \tag{4}$$

Decay constant λ is defined $\lambda = \ln 2/t_{1/2}$ through half-life $t_{1/2}$. Hydrodynamic dispersion tensor $\mathbf{D}$ is calculated [7] from the local hydrodynamic dispersion tensor $\mathbf{D}_{loc}$ as

$$\mathbf{D} = \mathbf{D}_{rot}^{-1}\,\mathbf{D}_{loc}\,\mathbf{D}_{rot}, \quad \mathbf{D}_{loc} \equiv \begin{pmatrix} D_p + \sqrt{\mathbf{v}\cdot\mathbf{v}}\,\alpha_L & 0 \\ 0 & D_p + \sqrt{\mathbf{v}\cdot\mathbf{v}}\,\alpha_T \end{pmatrix} \tag{5}$$

with rotation tensor $\mathbf{D}_rot$

$$\mathbf{D}_{rot} \boxminus \begin{pmatrix} \cos\phi & -\sin\phi \\ -\sin\phi & \cos\phi \end{pmatrix} \quad \mathbf{D}_{rot}^{-1} \equiv \begin{pmatrix} \cos\phi & -\sin\phi \\ \sin\phi & \cos\phi \end{pmatrix} \quad \theta = \arctan\frac{v_y}{v_x} \tag{6}$$

Retardation coefficient R, that represents the ability of porous media to remove dissolved chemical from the liquid to the solid matrix, and the media interaction source $\partial\Upsilon/\partial t$ are defined as

$$R = 1 + \frac{\varrho_b\,K_d}{\epsilon} \qquad \frac{\partial}{\partial t}\Upsilon = (1-R)\frac{\partial}{\partial t}C \tag{7}$$

We seek the steady-state seepage velocity field and the long-term transient concentration distribution by assuming known concentration and velocity fields at time t_0 and the adjacent boundary conditions of the Dirichlet and Neumann type, respectively.

3 Solution procedure

3.1 Poisson reformulation of the general transport equation

The solution of the problem posed is based on the general transport equation defined on a fixed domain Ω with boundary Γ, standing for a reasonably broad spectra of mass, energy, momentum, and species transfer problems

$$\frac{\partial}{\partial t}[\,\varrho\,\mathcal{C}(\Phi)\,] + \nabla \cdot [\,\varrho\,\mathbf{v}\,\mathcal{C}(\Phi)\,] = -\nabla \cdot (-\mathbf{D}\,\nabla\Phi) + S \tag{8}$$

with ϱ, Φ, t, $\mathbf{v}$, $\mathbf{D}$, and S standing for density, transport variable, time, velocity, diffusion matrix and source, respectively. Scalar function $\mathcal{C}$ stands for possible more involved constitutive relations. The solution of the governing equation for the transport variable at the final time $t = t_0 + \Delta t$ is sought, where t_0 represents the initial time and Δt the positive time increment under assumption of known inital conditions and boundary conditions of the Dirichlet, Neumann and Robin type. The governing equation is first transformed by splitting of the diffusion matrix

$$\mathbf{D} = D\,\mathbf{I} + \mathbf{D}' \tag{9}$$

into constant isotropic part $D\,\mathbf{I}$, with $\mathbf{I}$ denoting identity matrix, and the remaining nonlinear anisotropic part $\mathbf{D}'$. The following Poisson equation can be formulated afterwards

$$\nabla^2\Phi = \nabla\cdot\boldsymbol{\Theta} + \theta$$

$$\theta = [\,\frac{\partial}{\partial t}(\varrho\,\mathcal{C}(\Phi)) - S\,]/D, \quad \boldsymbol{\Theta} = [\,\varrho\,\mathbf{v}\,\mathcal{C}(\Phi) - \mathbf{D}'\,\nabla\Phi\,]/D \tag{10}$$

The inhomogenous terms are expanded as

$$\theta \approx \hat{\theta} + \hat{\theta}_{,\Phi}\;(\Phi - \hat{\Phi}), \quad \boldsymbol{\Theta} \approx \hat{\boldsymbol{\Theta}} + \hat{\boldsymbol{\Theta}}_{,\Phi}\;(\Phi - \hat{\Phi}) \tag{11}$$

with 'hat' denoting value at previous iteration. The final form of the transformed equation is

$$[\,\nabla^2 - \nabla\cdot\hat{\boldsymbol{\Theta}}_{,\Phi} - \hat{\theta}_{,\Phi}\,]\,\Phi = \nabla\cdot\hat{\boldsymbol{\Theta}} - \nabla\cdot\hat{\boldsymbol{\Theta}}_{,\Phi}\;\hat{\Phi} + \hat{\theta} - \hat{\theta}_{,\Phi}\;\hat{\Phi} \tag{12}$$

3.2 Basic elements of the DRBEM solution procedure

The solution of the governing equations is made with the recently developed DRBEM solution framework for the general transport equation [12]. The framework is sufficiently broad for inclusion of the completely anisotropic, non-linear, and multiphase problems. Its main characteristics is in an unified form of the solution procedure, able to cope with complicated coupled transport phenomena in an ordered way. The described features give DRBEM similar flexibility for coping with the complicated constitutive relations like classical numerical approaches such as the finite volume method. The transformed governing equation is time-discretized in a fully implicit manner, i.e.

$$\frac{\partial}{\partial t}(\varrho\,\mathcal{C}(\Phi)) \approx \frac{\varrho\,\mathcal{C}(\Phi) - \varrho_0\,\mathcal{C}(\Phi_0)}{\Delta t}$$

Subscript 0 denotes value at the initial time $t = t_0$ and no subscript denotes value at time $t_0 + \Delta t$. The governing equation is space-discretized by weighting the time-discretized governing equation over the domain Ω by

the fundamental solution of the Laplace equation T^*. Let us focus on two-dimensional (described in Cartesian coordinates p_x, p_y with base vectors $\mathbf{i}_x$, $\mathbf{i}_y$) situations

$$T^*(\mathbf{p};\mathbf{s}) = \frac{1}{2\,\pi}\,\log\frac{r_0}{r} \tag{13}$$

with $r^2 = (p_x - s_x)^2 + (p_y - s_y)^2$ and r_0 standing for the scaling constant. The following two integral equations can be deduced by assuming the properties of the fundamental solution and the Green's theorems

$$\int_\Gamma T^*\,\frac{\partial\Phi}{\partial n_\Gamma}\,d\Gamma - \int_\Gamma \frac{\partial T^*}{\partial n_\Gamma}\,\Phi\,d\Gamma - c_s^*\,\Phi_s = \int_\Omega [\,\nabla\cdot\boldsymbol{\Theta} + \theta\,]\,T^*\,d\Omega \tag{14}$$

$$\int_\Gamma \nabla T^*\,\frac{\partial\Phi}{\partial n_\Gamma}\,d\Gamma - \int_\Gamma \nabla\frac{\partial T^*}{\partial n_\Gamma}\,\Phi\,d\Gamma + \nabla(c_s^*\,\Phi_s) = \int_\Omega [\,\nabla\cdot\boldsymbol{\Theta} + \theta\,]\,\nabla T^*\,d\Omega \tag{15}$$

with $c^*(\mathbf{s})$ standing for the fundamental solution related coefficient. The right hand sides of the integral equations (14,15) are, for shorting the notation, written back in unexpanded form. The integral equation (14) is used for calculation of the unknown Φ and the integral equation (15) is used for calculation of the partial derivatives of Φ: $\Phi_{,\xi}\,;\xi = x, y$. The integral types that arise from defined weighting can be classified into the following six types

$$\int_\Omega \nabla^2\mathcal{F}\,T^*\,d\Omega, \quad \int_\Omega \mathcal{F}\,T^*\,d\Omega, \quad \int_\Omega \nabla\cdot\boldsymbol{\mathcal{G}}\,T^*\,d\Omega \tag{16}$$

$$\int_\Omega \nabla^2\mathcal{F}\,\nabla T^*\,d\Omega, \quad \int_\Omega \mathcal{F}\,\nabla T^*\,d\Omega, \quad \int_\Omega \nabla\cdot\boldsymbol{\mathcal{G}}\,\nabla T^*\,d\Omega \tag{17}$$

where $\mathcal{F}$ and $\boldsymbol{\mathcal{G}}$ refer to scalar and vector valued functions. They are computed as follows. Boundary geometry is approximated by N_Γ straight line segments, and spatial variation of the fields on each of the boundary segments is represented by constant interpolation functions with gridpoints coinciding with the geometrical centers of the straight line segments. The spatial variation of the fields in the domain is approximated by N_Ω global interpolation functions of the form

$$\mathcal{F}(\mathbf{p}) \approx \psi_u(\mathbf{p})\,\varsigma_u;\;\; u = 1, 2, \cdots, N+3, N = N_\Gamma + N_\Omega$$

The two-dimensional scaled augmented thin plate splines are used in this work (they do not require any free parameter)

$$\psi_n(\mathbf{p}) = r_n^2\,\log r_n;\;\; n = 1, 2, \cdots, N$$

$$\psi_{N+1}(\mathbf{p}) = p_x - p_x^0,\;\; \psi_{N+2}(\mathbf{p}) = p_y - p_y^0,\;\; \psi_{N+3}(\mathbf{p}) = 1 \tag{18}$$

with

$$r_n^2 = (\mathbf{p} - \mathbf{p}_{\underline{n}})\cdot(\mathbf{p} - \mathbf{p}_{\underline{n}}) \tag{19}$$

Scaling constants p_x^0 and p_y^0 stand for the mean coordinates of the domain Ω. The domain integrals are transformed into finite series of boundary integrals by using the dual reciprocity transformation. The elements of the involved boundary integral matrices and their derivatives are evaluated analytically. The application of the discretization and the described boundary conditions gives square system of N linear equations for solving Φ in the domain points and Φ or $\partial\Phi/\partial n_\Gamma$ in the boundary points. The involved systems af algebraic equations are solved, except for the pressure and pressure correction equations, by using GMRES technique with Jacobi preconditioning [9]. In case of the two pressure equations which are singular due to the prescribed Neumann boundary conditions over the whole boundary Γ, the systems are solved by the Householder reduction to bidiagonal form and QR diagonalization with shifts. All details of the formation of the systems of algebraic equations from the discretized equation and discretized initial and boundary conditions can be found in [12].

3.3 Coupled problem strategy and iteration margins

The four governing equations (pressure correction Poisson equation replaces the mass conservation equation) are in each timestep solved in an iterative bundle. Their representation within general transport equation context is shown in Appendix. Both species diffusion equations depend on the mass and momentum transport equations throug the velocity field. First, new pressure field is solved, based on the old velocity, pressure and temperature fields. Momentum equation is solved afterwards. Subsequently, pressure correction field is solved based on the new velocity field. The new velocity field is corrected through the pressure correction field. After convergence of the velocity and pressure fields at each timestep, the first species conservation equation is solved for concentration 1, followed by the species conservation equation for concentration 2. After each solution of any scalar Φ a relaxation with coefficient c_{rel} is made $\Phi = \hat{\Phi} + c_{\mathrm{rel}}\,(\,\Phi - \hat{\Phi}\,)$ and the timestep iterations are stopped when the criterion $(|\Phi_{\mathrm{avg}}| - |\hat{\Phi}_{\mathrm{avg}}|)/|\hat{\Phi}_{\mathrm{avg}}| < \epsilon_{\mathrm{itr}}$ is reached, where subscript avg represents the average gridpoint value. The criterion of reaching the steady state is $(|\Phi_{\mathrm{avg}}| - |\Phi_{\mathrm{avg}\,0}|)/|\Phi_{\mathrm{avg}\,0}| < \epsilon_{\mathrm{sts}}$.

3.4 DRBEM solution procedure verification

The DRBEM solution procedure has been verified for Navier-Stokes equations [10] by comparison with the classical Ghia-Ghia-Shin driven cavity test case. The extensive tests [11,12] for proper natural convection flow in porous media have been performed by comparison with the fine-mesh finite volume results. The convective-diffusive species transport, including the decay chain effects, has been checked by comparison with the CXTFIT code [13] results.

4. Simulation of the radionuclide transport

Two-dimensional test case, representing the transport of sorbing species from the repository to groundwater, is considered in the present paper. The calculated area with all involved materials and dimensions can be seen in Figure 1. The complete design calculation of the repository takes into account the following radionuclides: Co-60, Sr-90, Cs-137, Ni-59, Nb-94, I-129, H-3, and decay chains of U-238 (U-234, Th-230, Ra-226), and Pu-241 (Np-237, U-233, Th-229). Present test takes into account only the Pu-241 chain in which only Np-237 and U-233 have been considered. This can be justified by the half-life of Pu-241 (14.4 year) which is negligible in comparison with Np-237 and U-233 half lives and the Th-229 transport has been omitted due to limited conference paper scope. The design assumption of the unit thickness repository activity of Pu-241 is considered to be 31.6 Bq/m which corresponds to $5.1 \cdot 10^{-9}$ moles/m of Pu-241. Since the Pu-241 is assumed to be transformed to Np-237 instantaneously, this gives for a repository of crossection 104.4 m^2 initial concentration of Np-237: $C_1 = 0.0488\,mol/m^3$. The flow field in the surrounding geological structures and in the repository has been calculated under assuption of Dirichlet boundary conditions with seepage velocity $v_y = -0.25$ m/year on Γ. Because the velocity field is steady and not influenced by the concentrations, the velocity and transport calculations can be made decoupled. Concentration boundary conditions on the north boundary are of the Dirichlet type with $C_1 = C_2 = 0\,mol/m^3$ and other boundary conditions are of the Neumann closed system type. Coarse grid is applied with intention to show the robustness of the metod used. The whole domain is modelled (instead of the possible symmetrical half) to check the balance of the results.

properties	symbol	unit	soil	concrete	sediment
bulk density	ϱ_b	kg/m^3	2430	2590	2300
porosity	ϵ	1	0.1	0.12	0.15
permeability	$\mathcal{K}$	m^2	10 e−6	10 e−9	10 e−5
Np-237 $t_{1/2}$=2.14 e-6 years			molar activity: 6.16 e+9 Bq/mole		
distribution coefficient	K_d	m^3/kg	0.5	3.2	0.5
longitudinal dispersivity	α_L	m	1.00	0.10	0.40
transversal dispersivity	α_T	m	0.1	0.01	0.04
diffusion coeffisient	D_p	m^2/s	0.063	0.014	0.063
U-233 $t_{1/2}$=1.59 e−5 years			molar activity: 8.32 e+10 Bq/mole		
distribution coefficient	K_d	m^3/kg	0.035	0.79	0.035
longitudinal dispersivity	α_L	m	1.00	0.10	0.40
transversal dispersivity	α_T	m	0.1	0.01	0.04
diffusion coefficient	D_p	m^2/s	0.063	0.014	0.063

Table 1: Material properties [14].

The calculated seepage velocity field is shown in Figure 2, and the calulated temporal evolution of molar concentration and activity concentration of treated isotopes in point $p_x = 0\,\mathrm{m}, p_y = 5\,\mathrm{m}$ (boundary of the engineering barrier) are shown in Figures 3 and 4.

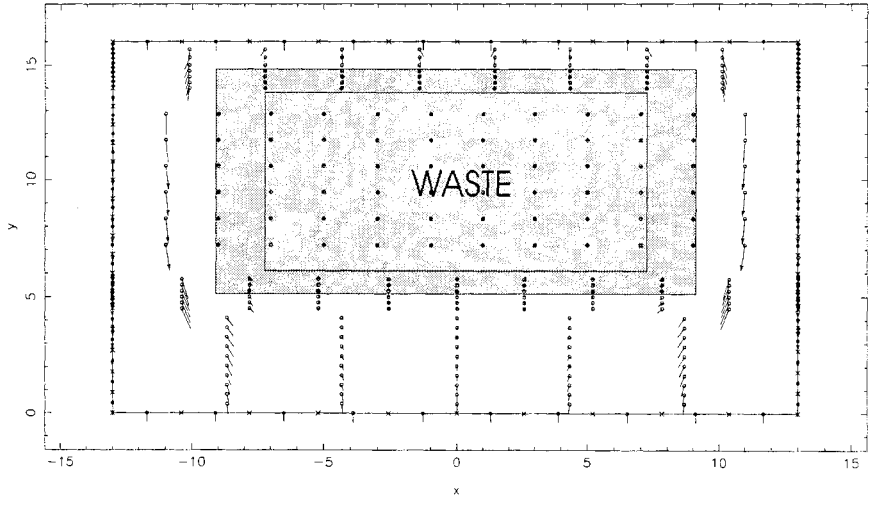

Figure 2: Solution of the velocity field for the proposed test.

5 Conclusions

This paper demonstrates the first attempt of PA calculation of the involved transport phenomena of sorbing and decaying solute in the geosphere by the dual reciprocity boundary element method. The calculations are made within the very recently developed standard computational framework for solution of the coupled transport phenomena by this method. This framework allows for coping with complicated coupled transport phenomena in an ordered way. The framework is sufficiently broad for inclusion of the completely anisotropic, non-linear, and multiphase problems and gives the method similar flexibility to handle complicated physics like the more established finite volume method. The principal advantage of the method used represents the boundary-only evaluation of all involved weighting boundary-domain integrals. Very simple test geometrical arrangement is used in this paper so that the results can be compared to other solutions (respective journal paper is underway). However, the boundary element - domain collocation point character of the method allows for easy treatment of much more involved geometries, very often found in realistic geosphere. This fact represents the main comparative advantage of this method with respect to the classical polygon-based computational approaches.

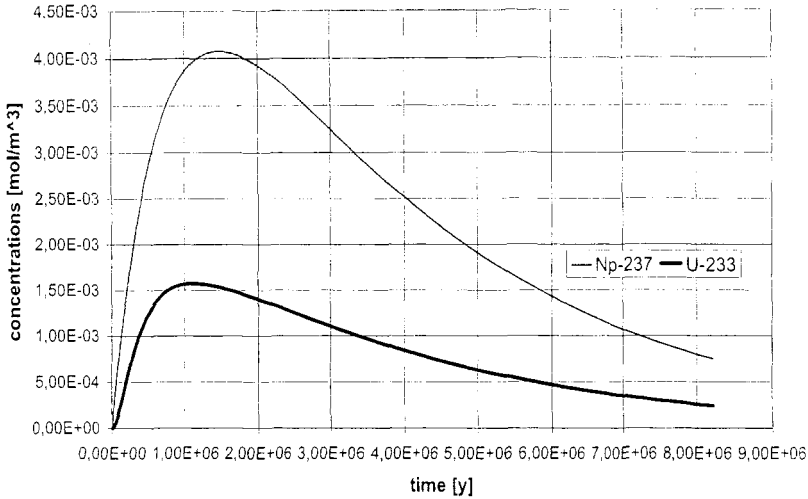

Figure 3: Molar concentration vs. time for Np-237 and its daughter U-233.

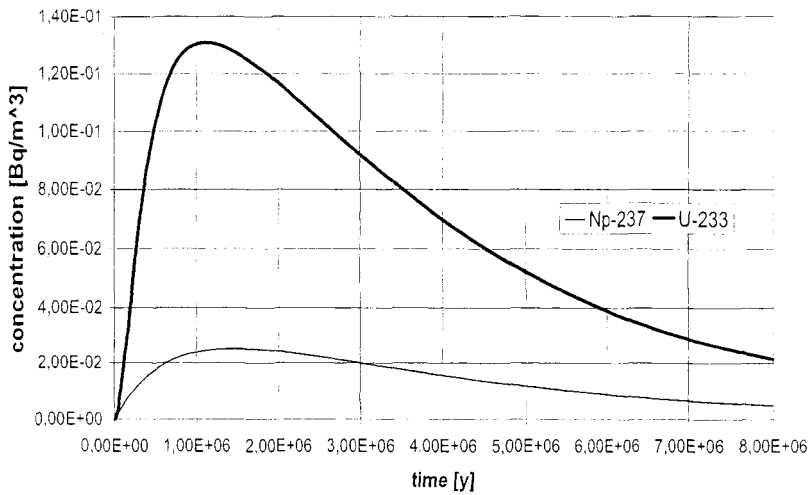

Figure 4: Activity concentration vs. time for Np-237 and its daughter U-233.

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