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Numerical simulation of radionuclides migration in the far field of a geological repository

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Abstract

Safety conditions associated with geological repositories must be guaranteed also in the case of radionuclides migrating from the near field to the far field of a geological repository and to the external environment. For this reason the migration process of radionuclides and the factors affecting the process patterns have a crucial importance. In the present article, in order to simulate the migration process of radionuclides in the far-field of a geological repository, the groundwater simulation code PMWIN (Processing Modflow) is used, following a methodology applied by the same authors in a previous work. The present case study refers to a non-uniform groundwater flow field and shows the influence of two important parameters, the distribution coefficient and the hydraulic gradient. The results are compared with the ones previously obtained for the uniform flow case and the different scenarios are discussed.

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Keywords: radionuclides migration; far-field; numerical model; geological repositories

1. Introduction

In the last decades in many countries significant progresses have been made in geological repository R&D with the aim to solve the problem of a safe and acceptable solution for existing and future inventories of high-activity, long-lived radioactive waste. Worldwide, whatever is the existing nuclear fuel cycle, the geological repository is considered to be the better final solution. In Europe, Finland and Sweden have completed their own repository design, selected the sites and now the repositories are in the construction phase. Both disposals will be likely to be operative by 2025. In France and in Switzerland studies for the project development and site identification are progressing and it is expected that the facilities will be in operation by 2030. Several other countries as Argentina, Australia, Japan, Republic of

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Korea, Russia are carrying out similar experiences. The improvements in the "closed fuel cycle" studies and, in particular, in P&T (Partitioning and Transmutation) processes will lead to an optimization of the performances of the geological disposal mainly in terms of :

- reduction of the potential source of radiotoxicity to a factor larger than 100 (both of the mass of transuranic elements and of their radiotoxicity) [1],
- reduction of the size of the storage because of the elimination (from the total amount of wastes) of some long-lived FP (mainly Cs and Sr) which are responsible of approximately two-thirds of the total heat decay.

In this context too, deep geological disposal [2] [3] remains the preferred solution for HLW and long-lived radioactive waste.

2. Goal of the study

Actinide ions can be transported through porous media also adsorbed by colloids. In this case, not only the nature of the colloids binding to actinide ions but also the size and the stability of the colloids are important parameters that determine the relevance of the radionuclide migration. Disregarding the aspects related to colloid-mediated radionuclide migration, the present study focuses on numerical investigations carried out using MODFLOW/MT3DMS multispecies transport.

The present study is the continuation of a previous one [4] where the groundwater flow was considered to be horizontal and uniform. The geometry and the discretization of the flow domain, the input parameters, the initial conditions and the scenarios are the same, as defined in the previous one. Only the boundary conditions are different. Here the flow is not uniform, it has horizontal and vertical components.

Scope of the present study is to outline through the analysis of the scenarios the relevance of the basic mechanisms influencing the radionuclides migration as the advection component related to the hydraulic gradients and the retardation component due to the adsorption process.

Scope of the present study is also to evaluate the influence of the cells size on the results precision.

3. Mathematical Approach

Nomenclature

c	concentration in the fluid [M/L^3]
c_a	concentration in the solid matrix
D	dispersion coefficient [L^2/T]
D_o	diffusion coefficient [L^2/T]
h	piezometer level [L]
K_{ij}	tensor of the hydraulic conductibility [L/T]
K_d	distribution (partition) coefficient [L^3/M]
n	porosity
Q	source term [1/T]
S	specific yield [1/L]
$T_{1/2}$	half-life of radionuclide [1/T]
v	velocity [L/T]
α	dispersivity [L]
λ	decay constant [1/T]
ρ	density [M/L^3]

The two main equations are the groundwater flow equation and the groundwater transport equation.

The groundwater flow equation is derived from the continuity equation and from Darcy's equation. If the medium is homogeneous and isotropic, the groundwater equation can be written as follows:

$$\frac{\partial}{\partial x_i}\left(K_{ij}\frac{\partial h}{\partial x_j}\right) = S\frac{\partial h}{\partial t} \pm Q \tag{1}$$

Where i and j are the principal directions, K is the tensor of the hydraulic conductibility, h is the piezometer level, S is the specific yield, Q is the source/sink term for a unit volume and a unit time.
The groundwater transport equation is composed by several terms and can be written as follows:

$$\frac{\partial c}{\partial t} + \frac{(1-n)}{n}\rho_s\frac{\partial c_a}{\partial t} = -\frac{\partial}{\partial x_i}(v_i c) + \frac{\partial}{\partial x_i}\left(D_{ij}\frac{\partial c}{\partial x_i}\right) - \lambda c - \sum Qc_{in} \tag{2}$$

(I) (IV) (II) (III) (V) (VI)

where C is the concentration, n is the porosity, ρ is the density, v is the velocity, D is the dispersion coefficient, λ is the decay constant, Q is the source /sink term.
Term I represents the variation of the concentration of the contaminant with time.
Term II describes the advective transport. Term III represents the process of hydrodynamic dispersion and it is the result of 2 phenomena, the molecular diffusion and the mechanical dispersion.
The molecular diffusion occurs if there is a concentration gradient. In a porous medium the molecular diffusion is described by the Fick's law as in the free water, but instead of the coefficient of molecular diffusion D_o (order of magnitude of 10^{-9} m^2/s) a reduced coefficient, called effective coefficient, is used.
The mechanical dispersion is due to three main phenomena related to the friction at the pore walls, to the tortuosity of the pathlines and to the velocity gradients within the pores.
For an isotropic medium the components of mechanical dispersion D can be expressed as a function of the water velocity v and of 3 parameters, the longitudinal dispersivity α_L, the transversal dispersivity α_T, and the vertical dispersivity α_v, namely:

D_L= v α_L where α_L=10% x
D_T= v α_T where α_T=10% α_L
D_v= v a_v where α_v=1% α_L

and x is the lenght of the plume.
Term IV represents the adsorption. It causes a decrease of the concentration of the radionuclide in the liquid phase and the delayed transport of the radionuclide compared to the water flow. If the process ofsorption is faster than the advective process, equilibrium is reached between the concentration c_a in the solid matrix and the concentration c in the fluid.
If the relationship between the two concentration values at equilibrium and at constant temperature is linear, c_a can be expressed as a function of the partition coefficient K_d, namely:

$$c_a = K_d c \tag{3}$$

Term V in the transport equation represents the decay process and λ is the decay rate. This can be expressed as a function of the half-life of the radionuclide $T_{1/2}$ as follows (first order kinetics):

$$\lambda = (ln2)/T_{1/2} \tag{4}$$

Term VI is the sink/source term.

4. Numerical simulation

A mathematical model is built using PMWIN (Processing Modflow) [5] [6], a modular three-dimensional finite-difference groundwater model of the U.S. Geological Survey.

The numerical example simulate the migration process of radionuclides (Pu 239) in a confined aquifer, 100 m thick, homogeneous and isotropic, confined between two horizontal impermeable layers, deep below the topographic level. The groundwater flow at the west boundary is uniform and horizontal. Groundwater flows from the west boundary towards a sink point located at the upper corner at the east boundary. The sink point is the outflow from the model domain and in nature can be the bottom of a well, a fracture, or any connection to earth surface above. This situation (Fig.1 (left)) generates a potential gradient in both horizontal and vertical directions. A 2-dimensional vertical model is built. The flow domain is discretized with 100 x 100 cells and each cell is 1m x 1m. The thickness of the model is 1m and the cell volume is 1 m3. In Fig.1 (right) the piezometer lines and the flow paths are shown

The flow domain has 3 impermeable boundaries: the upper boundary, the lower boundary and the east boundary. The west boundaries is a constant head boundary. The upper right corner is a sink point (for instance the bottom of a well or a fracture). The difference in constant head ΔH between the west boundary and the sink point generates the hydraulic gradient and the advective flow.

In the numerical example the hydraulic conductivity of the aquifer K is 0.05 m/y, the porosity n is 0.1.

The decay rate λ of Pu 239 is 2.8755 10^{-5}.

The repository (20 m long and 10 m high) is simulated as a source with constant concentration equal to the maximum concentration at solubility of Pu 239 equal to .00024 kg/m^3.The effective molecular diffusion coefficient is 0.015768 m^2/y. The longitudinal dispersivity is assumed to be 2 m and the vertical dispersivity 0.02 m. The bulk density of the soil is 1480 kg/m^3.

In order to quantify the opposite effects of the hydraulic gradients in the groundwater regimes and of the adsorption in the soil, 4 scenarios are presented here, based on 2 values of the hydraulic head drop (H=20 m and H=40 m) and 2 values of the partition coefficient (K_d =0.003 and K_d= 0.006), namely:

Scenario 1:	H=20 m	K_d=0.003
Scenario 2:	H=20 m	K_d=0.006
Scenario 3:	H=40 m	K_d=0.003
Scenario 4:	H=40 m	K_d=0.006

The solution scheme for the transport simulation is the upstream finite difference method (upstream weighting scheme) and the courant number is equal to 0.75.

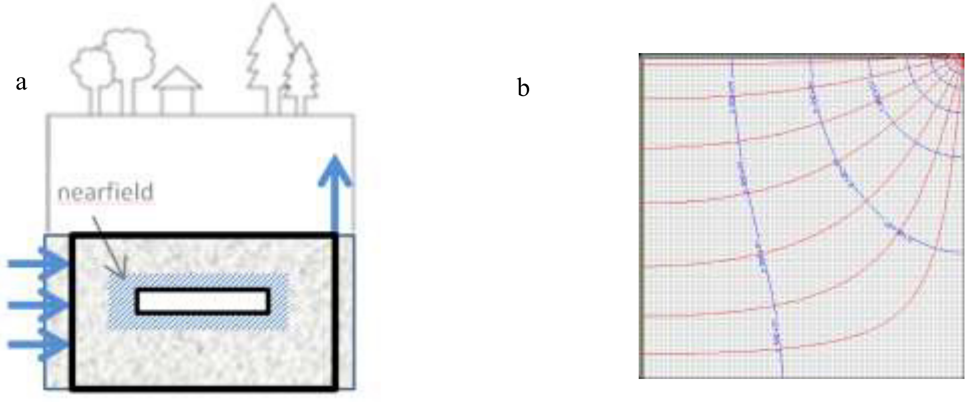

Fig.1 The location of the geological repository (a) and the groundwater flow domain (b)

5. Results

The results of the numerical simulations are summarized below. The spatial distribution of the concentration values at t=5000 years are shown in Fig.2 (scenario 1 and 2) and Fig.3 (scenario 3 and 4).

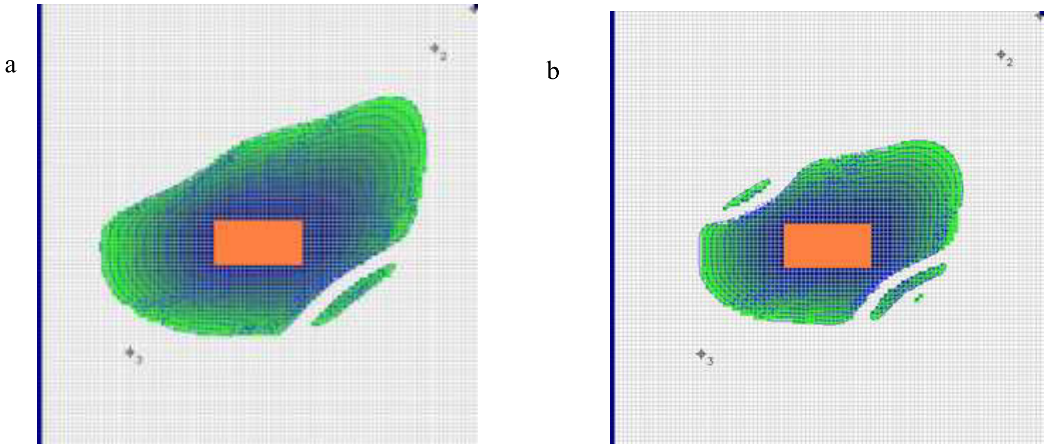

Fig. 2. Spatial distribution of the concentration values ranging from 0.00024 kg/m^3 to 10^{-14} kg/m^3 (Simulation results with cell size 1x1m): Scenario 1 (a) and Scenario 2 (b)

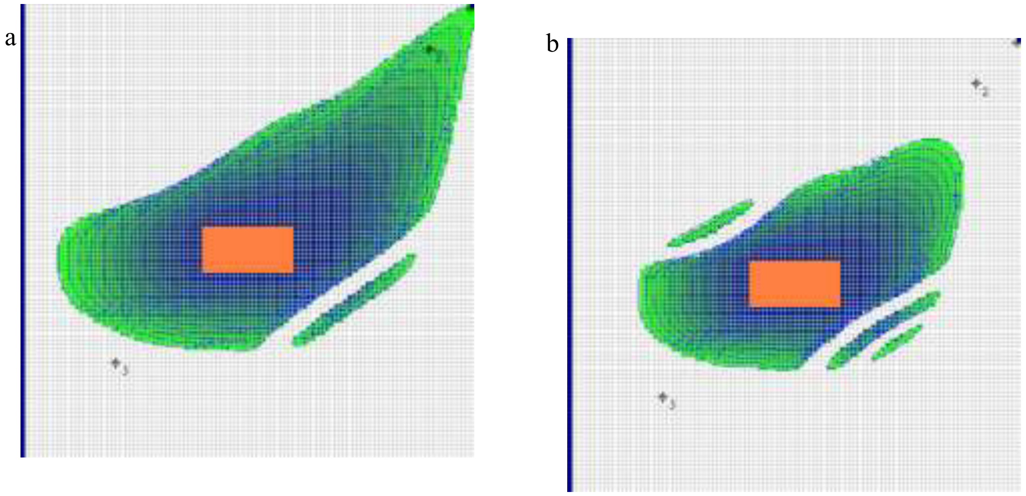

Fig. 3. Spatial distribution of the concentration values ranging from 0.00024 kg/m^3 to 10^{-14} kg/m^3: Simulation results with cell size 1x1m): Scenario 3 (left) and Scenario 4 (right)

As in the previous study where it was considered the case of the horizontal uniform flow, the smallest contamination plume corresponds to scenario 2 (low hydraulic gradient and high distribution coefficient) and the biggest contamination plume corresponds to scenario 3 (high hydraulic gradient and low distribution coefficient). The spatial distribution of the concentration values for scenario 1 and scenario 4 are not similar but the longitudinal dimension is comparable. In fact, the plume extends in both cases approximately 38 m forward and 25 m backward.

The effects of higher hydraulic gradients compensate the effects related to higher distribution coefficients (higher retardation). It has to be noticed that in all four scenarios the solutions are affected by numerical errors, which are visible along the largest contours, which correspond to the lowest concentration values. There the numerical solutions show some oscillations around the zero in the transversal direction of the plume direction. This aspect is more evident for scenario 4, which corresponds to the simulation with a higher hydraulic gradient and higher distribution coefficients. This problem is more evident at the lower right corner of the flow domain where the curvature of the path lines is bigger.

The 4 simulations were repeated with a finer discretization: thee flow domain is discretized with 200 x 200 cells and the cell size is 0.5m x 0.5m. Fig.4 and fig.5 show the results with the finer grid. It can be seen that the oscillations of the numerical solution have been reduced.

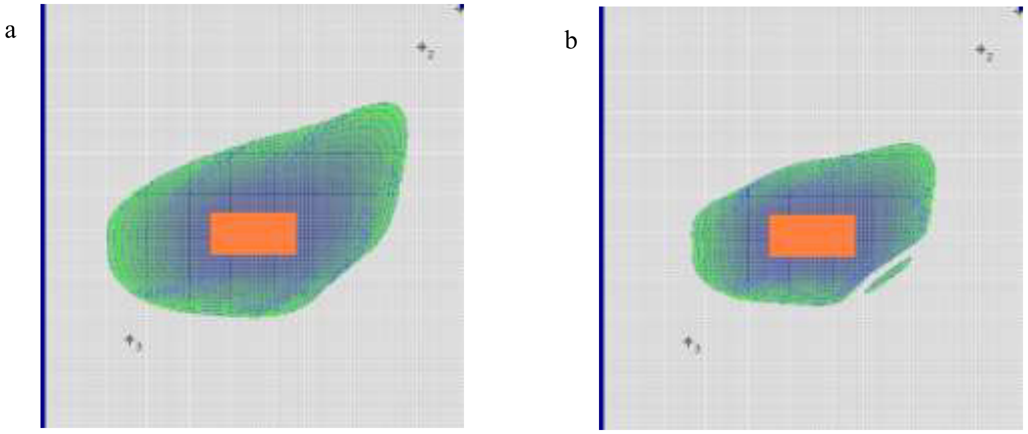

Fig. 4. Spatial distribution of the concentration values ranging from 0.00024 kg/m^3 to 10^{-14} kg/m^3: (Simulation results with cell size 0.5x0.5m): Scenario 3 (a) and Scenario 4 (b)

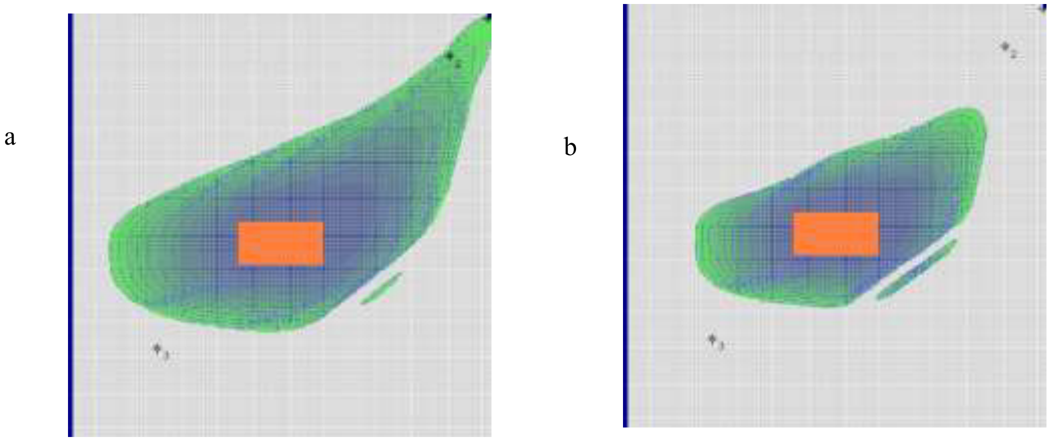

Fig. 5. Spatial distribution of the concentration values ranging from 0.00024 kg/m^3 to 10^{-14} kg/m^3: (Simulation results with cell size 0.5x0.5m): Scenario 3 (a) and Scenario 4 (b)

6. Conclusions

The simulation of the migration process in the far-field of a geological repository is an important issue in the safety assessment of the geological repository. Through the definition and the analysis of a set of scenarios it was possible to outline the role of the hydraulic gradient and of the distribution coefficient on the migration process, in particular on the longitudinal and transversal extension of the plume. An appropriate grid refinement can reduce the numerical errors, which are more evident in the zones where the pathlines curve from the horizontal direction to the vertical direction.

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Biography

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