PROCESS-LEVEL MODELLING RELATED TO GEOLOGICAL DISPOSAL OF THE SPENT NUCLEAR FUEL

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Abstract

In this paper, the concentration profiles in a bentonite block calculated by the simulation tool GoldSim were verified through comparison to an analytical solution of non-steady state diffusion equation which was derived based on the certain initial and boundary conditions. The impact of bentonite buffer discretization on the release rates (from the near field) of modelled radionuclides was also evaluated. The size of discretization has influenced mainly the release rate of ^{242}Pu which is highly sorptive radionuclide. In this case the simulation tool GoldSim was also used.

1. Introduction

The geological repository is based on the principle of multi-barrier concept (combination of engineering barriers and a suitable geological formation). The typical components of the engineered barrier system (EBS) are the very waste form (e.g. spent nuclear fuel), disposal container and buffer material. Buffer and backfill materials are designed to fill the empty spaces after excavation. In many international concepts of geological disposal, a bentonite as buffer and backfill material is proposed. Diffusion is considered to be a dominant mechanism regulating the transport of contaminants through the bentonite buffer. Process-level models are developed in order to obtain a thorough knowledge about a part of the disposal system or about a particular process that can affect the disposal system or its part.

2. Diffusion theory

Diffusion occurs when radionuclides migrate from a region of higher concentration to a region of lower concentration, even in the absence of the groundwater flow. Diffusion is caused by the random motion of molecules and atoms (Brownian motion) and the diffusion flux is proportional to the concentration gradient [1]. Diffusion is described by Fick's laws, whereby the First Fick's law applies to the diffusion flux under steady state conditions and the Second Fick's law applies to the systems where the concentration gradient $\partial C/\partial x$ [kg/m^3/m] is changing with time (non-steady state conditions).

Under the non-steady state conditions, the one-dimensional equation is derived from the law of the conservation of mass. In case of sorptive elements, a change in the solute concentration is also influenced by a change in the sorbed concentration. When the porosity ε [-], the effective diffusion coefficient D_e[m^2/s] and the dry density ρ_d [m^3/kg] are assumed to be constant, the Second Fick's law(including the sorption)can be expressed as [2]:

$$\varepsilon \frac{\partial C}{\partial t} = D_e \frac{\partial^2 C}{\partial x^2} - \rho_d \frac{\partial S}{\partial t} \quad (1)$$

where C [m^3/kg] is the solute concentration and S represents the sorbed concentration per unit mass of solid phase [kg/kg]. The simplest expression of equilibrium sorption is the linear sorption isotherm. For systems described by a linear isotherm, K_d[m^3/kg] is a constant and the sorbed concentration can be expressed by the simple relation $S = K_d.C$. Eq.(1) can be then rewritten as [2]:

$$\frac{\partial C}{\partial t} = \frac{D_e}{\varepsilon + \rho_d K_d} \frac{\partial^2 C}{\partial x^2} = D_a \frac{\partial^2 C}{\partial x^2} \quad (2)$$

D_a[m^2/s]is the apparent diffusion coefficient and the term $\varepsilon + \rho_d.K_d$ is also known as the capacity factor α[-].

3. GoldSim –simulation tool

Modelling was carried out using the simulation software GoldSim which RT (Radionuclide Transport) module allows users to dynamically model mass transport within a complex system of engineering and natural barriers. The basic mass balance equation for a cell i when a radioactive decay, mass flux links and a direct input of species into the cell i are included can be described by [3]:

$$m'_{is} = -m_{is}\lambda_s + \sum_{c=1}^{N_i} f_{cs} + S_{is} \quad (3)$$

where m'_{is}[kg/s] is the rate of mass change of species s in the cell i,m_{is}[kg] is the mass of species s in the cell i,λ_s[s^{-1}] is the decay rate for species s,N_i is the number of mass flux links from/to the cell i,f_{cs}[kg/s] is the flux of species s through the mass flux link $c$and S_{is}[kg/s] represents the rate of direct input of species s to the cell i from the external sources.

GoldSim uses a finite volume approach to solve the mass transport of radionuclides. The examined region is divided into small volumes in which the mass change is computed. The finite volumes are in the GoldSim called *cells*. If the solubility limit is not reached, the diffusive flux from the cell i into the cell j can be described by [3]:

$$f_{s,i\to j} = D_{s,i\to j}(C_{s,i} - C_{s,j}) \quad (4)$$

where $D_{s,i\to j}$ [m^3/s] is the diffusive conductance for species s in the $i \to j$ direction,$C_{s,i}$[kg/m^3] is the dissolved concentration of species s inthe cell i and $C_{s.j}$[kg/m^3] represents the dissolved concentration of species s in the cell j. When the properties of a solid material in the individual cells are the same and species are transported only by single fluid, the diffusive conductance between the cells can be expressed as $D_s=(A.\varepsilon.\tau.D)/l$, where A[m^2] is the diffusive area, D [m^2/s] is the diffusion coefficient, τ[-] is the tortuosity and l[m] is the sum of diffusive lengths in the cell i and j. The sorption of the species s in cells is assumed to happen instantaneously.

4. Diffusion through the bentonite block

The aim of these calculations is to compare results of concentration profiles in the bentonite block calculated using the simulation tool GoldSim with the analytical solution of

the non-steady state diffusion equation derived based on the certain initial and boundary conditions.

4.1 Analytical solution – constant boundary concentrations

The concentration profiles were calculated for the bentonite block of thickness L [m] and following initial and boundary conditions:

$C = C_1, x = 0, t \geq 0,$

$C = 0, x = L, t \geq 0,$

$C = 0, 0 < x < L, t = 0.$

Based on the initial and boundary conditions, it is possible to see that there is initially no concentration of radionuclide in the bentonite block and the in-boundary concentration is considered to be constant (C_1), whereby the out-boundary concentration is kept zero.The concentration profile is expressed in the form of trigonometrical series (obtained by the method of separation of variables and by applying the initial and boundary conditions) and is defined by [4]:

$$\frac{C}{C_1} = 1 - \frac{x}{L} - \frac{2}{\pi}\sum_{n=1}^{\infty}\frac{1}{n} sin\left[\frac{n\pi x}{L}\right] exp\left[\frac{-n^2\pi^2 D_a t}{L^2}\right] \quad (5)$$

After a sufficient time (when steady state conditions are reached), the exponential term is diminished and the concentration profile becomes a straight line.

4.2 Comparison of the results obtained by analytical and numerical method

The calculations were performed for a fictive stable nuclideanddifferent distribution coefficients–0 m^3/kg, 0.001 m^3/kg, 0.1 m^3/kg and 1 m^3/kg. Other transport and medium properties were not changed and no solubility limits were assumed in the individual calculation cases: De = 7x10^{-12} m^2/s, ε=0.36, ρ_d= 1600 m^3/kg, A = 12.05 m^2,L = 0.3 m. To eliminate the discretization error, the bentonite block was discretized into 20 and 60 cells and the concentration profiles between these two discretization cases were also compared. The concentration profiles were evaluated in the time t = 200 000 days.

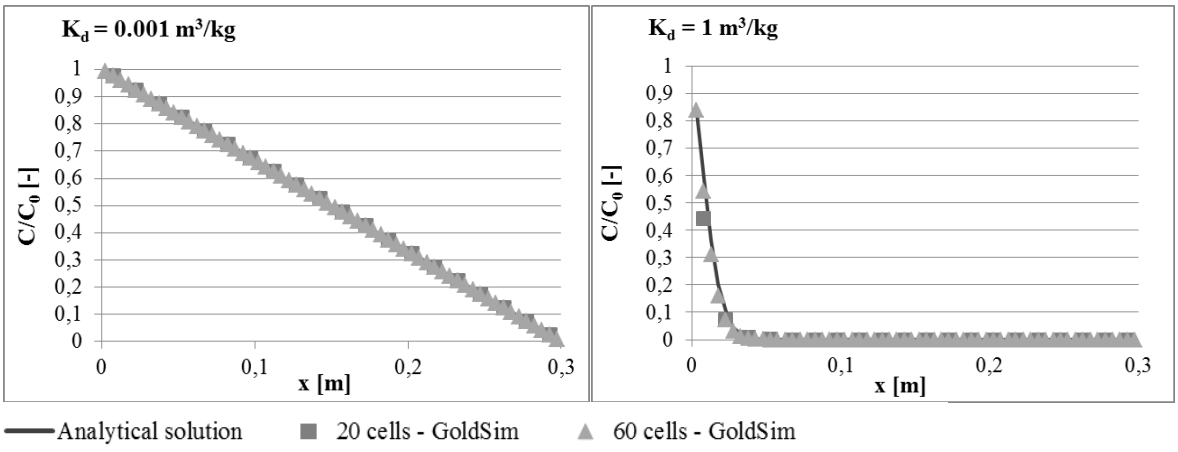

Fig.1:*Comparison of concentration profiles of stable nuclide with two different distribution coefficients.*

The concentration profiles of the radionuclide with different distribution coefficients are shown in Fig. 1. It can be seen that the results of concentration profiles calculated by analytical method and by using the GoldSim simulation tool (finite volume approach) are in very good agreement. In the first two cases (when K_d is equal to 0 m_3/kg and 0.001 m3/kg), the steady state conditions were reached and the concentration profiles became the straight lines.

5. Diffusion through the cylindrical buffer

As the GoldSim uses a finite volume approach to calculate the mass transport of radionuclides through a net of cells, these calculations are aimed at determination of impact of buffer discretization on release rates from the bentonite buffer.

It is considered that the disposal container is surrounded by the bentonite buffer with a wall thickness of 0.3 m. Disposal container fails after 1000 years and radionuclides are released congruently with the dissolution of the fuel matrix. The radionuclides migrate through the buffer by radial diffusion (angular or axial dependency is not considered). The cylindrical buffer was discretized into 1, 5, 15 and 30 cells which explicitly represent the finite volumes. Three representative nuclides were chosen to evaluate the impact of the buffer discretization on individual release rates - ^{226}Ra, ^{36}Cl and ^{242}Pu. ^{36}Cl is a very mobile radionuclide with poor retentive properties (no sorption and no solubility limit was considered in the calculations). On the other hand, ^{242}Pu is very sorptive radionuclide and^{226}Ra is highly limited by its solubility. The solubility limit of ^{242}Pu was conservatively set to -1 mol/m^3(which is not a realistic assumption) which means that a nuclide is infinitely soluble in the near field of the repository. This assumption was done to evaluate a release rate of a nuclide with no solubility limitation and high distribution coefficient. For ^{36}Cl 12 % IRF (instant release fraction) was considered. IRF is a fraction of inventory which is after water contact released rapidly, in the term of long-term safety instantaneously. An overview of these properties can be found in Tab. 1.

Tab. 1:*Basic properties of modelled radionuclides.*

Species	T [yr]	D_e [m^2/s]	Kd [m^3/kg]	Solubility [mol/m^3]	IRF [%]
^{242}Pu	3.75x10^5	3x10^{-12}	20	-1	0
^{36}Cl	3.01x10^5	2x10^{-10}	0	-1	0
^{226}Ra	1.60x10^3	2x10^{-10}	0.002	2x10^{-8}	12

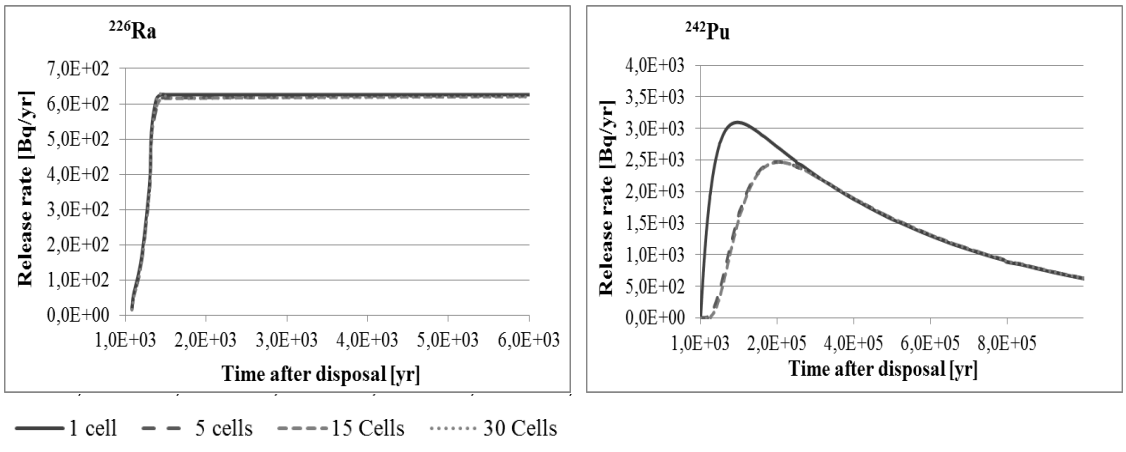

Fig.2:*Comparison of release rates indifferent discretization cases (^{226}Ra and ^{242}Pu).*

Based on the results it can be seen that significant differences in the release rates have occurred only in cases when the bentonite buffer was represented by a single cell. Representation of the buffer by one cell has the greatest impact on the release rate of ^{242}Pu (Fig. 2), which has the highest distribution coefficient (higly sorptive radionuclide). The different discretization has almost no impact on the ^{226}Ra (Fig. 2) which is significantly limited by its solubility.A lower degree of discretization caused the underestimation of the release rates of ^{36}Cl(with increasing degree of discretization, differences in the release rate was decreasing). On the other hand the release rate of ^{242}Pu was overestimated.

6. Conclusion

Based on the results it can be concluded that the concentration profiles of the stable radionuclide with specified different distribution coefficients calculated by the analytical method and by using the simulation tool GoldSim are in very good agreement. In the case when K_d = 0 m^3/kg and K_d = 0.001 m^3/kg, the simulation time (t = 200 000 days) was sufficient to reach the steady state conditions and the concentration profiles became the straight lines.

The impact of buffer discretization on release rates of modelled species was also evaluated. Significant differences in the release rates have occurred only in cases where the bentonite buffer was represented by a single cell. It can be said that the discretization of the buffer into 5 cells is sufficient to obtain reasonable results. The size of discretization influenced mainly the release rate of ^{242}Pu which is highly sorptive radionuclide.

Acknowledgement

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References:

[1] United States Environmental Protection Agency. Important Processes Affecting the Fate and Transport of Organic Compounds in the Subsurface. [online]. [cit. 20. April 2016]. Available: <http://www.epa.gov/superfund/health/conmedia/gwdocs/pdfs/protoapb.pdf>.

[2] García-Gutiérrez, M., Cormenzana, J.L., Missana, T. et. al.: Overview of laboratory methods employed for obtaining diffusion coefficient in FEBEX compacted bentonite. Journal of Iberian Geology,Volume32, ISSN 1698-6180, p.37-53 (2006).

[3] GOLDSIM Technology Group LLC. GoldSim Contaminant Transport Module, User`s Guide. Washington, USA (2014).

[4] Crank, J.: Mathematics of diffusion. 2nd edition, ClarendonPress, Oxford (1975).