RADIONUCLIDE DIFFUSION IN A HOLLOW CYLINDRICAL GEOMETRY OF BENTONITE BUFFER

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Received 16 May 2017; accepted 25 May 2017

Abstract

This paper gives an analytical solution for a non-steady state diffusion equation in a hollow cylindrical geometry of bentonite buffer. The analytical solution was derived based on the certain boundary and initial conditions and assuming a process of linear sorption. Consequently, concentration profiles and diffusive outfluxes from the hollow cylindrical buffer calculated by the simulation tool GoldSim were verified through comparison to the analytical calculations performed by MATLAB. Within these calculations, the impact of the spatial and temporal discretization on the results of concentrations and outfluxes was also evaluated.

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. Buffer material, which immediately surrounds the disposal container, should provide favourable thermo-hydro-mechanical-chemical conditions for the disposal container, limits water access and retards the radionuclide movement. 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.

2. Diffusion equation

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. 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 is changing with time (non-steady state conditions). For the one-dimensional radial diffusion in the cylindrical coordinate system, the Second Fick's Law is given by [1]:

$$\frac{\partial C}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(rD\frac{\partial C}{\partial r}\right), \tag{1}$$

where $\partial C/\partial r$ [kg/m^3/m] is the concentration gradient and D is the diffusion coefficient [m^2/s]. In this case the concentration is only a function of radius r [m] and time t [s].

In porous media, such as a compacted bentonite, where the geometry complicates the diffusion, the pore diffusion coefficient D_p is introduced. The pore diffusion coefficient takes into account the tortuous paths of the moving particles, the tortuosity τ [-], and also the irregular form and size of the individual pores, constrictivity δ [-]. As the diffusion takes place only through the interconnected pores, the effective diffusion coefficient D_e is defined by [2]:

$$D_e = \varepsilon D_p = \varepsilon \frac{\delta}{\tau^2} D_w = \varepsilon G D_w \,, \tag{2}$$

where ε [-] is the porosity of the material, D_w represents the diffusion coefficient in free water and G [-] is the pore geometric factor.

In case of sorptive elements, a change in the solute concentration is also influenced by a change in the sorbed concentration. When the porosity, dry density ρ_d [m^3/kg] and effective diffusion coefficient of the material are assumed to be constant, the Second Fick's law (including the sorption) can be expressed as [1,2]:

$$\varepsilon \frac{\partial C}{\partial t} = D_e \frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial C}{\partial r}\right) - \rho_d \left(\frac{\partial S}{\partial t}\right). \tag{3}$$

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

$$\frac{\partial C}{\partial t} = \frac{D_e}{\varepsilon + \rho_d K_d} \frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial C}{\partial r}\right) = D_a \frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial C}{\partial r}\right). \tag{4}$$

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 – computational method

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. It is also possible to simulate a radioactive decay and ingrowth of individual radionuclides. The basic mass balance equation for a cell i when mass flux links and a direct input of species into the cell i are included can be described by Equation 5 [3]:

$$m'_{s,i} = \sum_{c=1}^{N_i} f_{cs} + S_{s,i} \,, \tag{5}$$

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

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 Equation 6 [3]:

$$f_{s,i\rightarrow j} = D_{s,i\rightarrow j}(C_{s,i} - C_{s,j}), \tag{6}$$

where $D_{s,i\rightarrow j}$ is the diffusive conductance for species s in the $i \rightarrow j$ direction [m^3/s], $C_{s,i}$ is the dissolved concentration of species s in the cell i [kg/m^3] and $C_{s,j}$ represents the dissolved concentration of species s in the cell j [kg/m^3]. 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 $D_s=(A.\varepsilon.\tau.D_w)/l$, where A is the diffusive area [m^2] and l is the sum of diffusive lengths in the cell i and j [m]. GoldSim's tortuosity τ is in this definition equal to pore geometric factor G.

When partition (distribution) coefficients between the various media are specified in the model, a contaminant mass is instantaneously distributed between the media based on the distribution coefficients and masses/volumes of the various media present [3].

GoldSim solves a matrix equation representing the coupled system of equations based on the backwards-difference (fully implicit) algorithm for each cell net to compute concentrations and fluxes. In order to enhance the accuracy of the backwards-difference method, GoldSim automatically divides every timestep into a number of fractional timesteps, depending on the user's selected precision settings (high – 10 fractional timesteps, medium – 4 fractional timesteps, low – 1 fractional timestep) [3].

4. Diffusion in a hollow cylinder – analytical solution for the transient state

Considering the hollow cylinder where there is initially no concentration of nuclides and the in-boundary concentration is considered to be constant (C_1), whereby the out-boundary concentration is kept zero (Fig. 1):

$C = C_1,\ r = a,\ t > 0,$

$C = 0,\ r = b,\ t > 0,$

$C = 0,\ a < r < b,\ t \le 0.$

Fig. 1. *Schematic illustration of diffusion in a hollow cylinder.*

By using the solution to the problem of diffusion in the hollow cylinder given by Carslaw and Jaeger and by applying the initial and boundary conditions, the concentration profile can be calculated as [1,2,4]:

$$\frac{C}{C_1} = -\frac{ln\left(\frac{r}{b}\right)}{ln\left(\frac{b}{a}\right)} + \pi \sum_{n=1}^{\infty} \frac{J_0^2(b\alpha_n)U_0(r\alpha_n)}{J_0^2(a\alpha_n) - J_0^2(b\alpha_n)} exp(-D_a \alpha_n^2 t), \tag{7}$$

where

$$U_0(r\alpha_n) = J_0(r\alpha_n)Y_0(a\alpha_n) - J_0(a\alpha_n)Y_0(r\alpha_n), \tag{8}$$

and

$$J_0(a\alpha_n)Y_0(b\alpha_n) - J_0(b\alpha_n)Y_0(a\alpha_n) = 0, \tag{9}$$

where J_0 and Y_0 are the Bessel function of the first and second kind and zero order, α_n are the positive roots of Equation 9 and a [m] and b [m] represent the inner and outer radius of the hollow cylindrical buffer. After a sufficient time, the exponential term is diminished and the steady state conditions are reached.

The flux of the transient state at the out-boundary J_t^b [kg/m^{-2}s^{-1}] can be calculated as [1]

$$J_t^b = -D_e \left(\frac{\partial C}{\partial r}\right)_{r=b}, \tag{10}$$

$$J_t^b = \frac{D_e C_1}{b\, ln\left(\frac{b}{a}\right)} - \pi C_1 D_e \sum_{n=1}^{\infty} \frac{\alpha_n J_0^2(b\alpha_n)[J_0(a\alpha_n)Y_1(b\alpha_n) - J_1(b\alpha_n)Y_0(a\alpha_n)]}{J_0^2(a\alpha_n) - J_0^2(b\alpha_n)} exp(-D_a \alpha_n^2 t), \tag{11}$$

where J_1 is the Bessel function of the first kind and Y_1 is the Bessel function of the second kind and first order.

4.1 Comparison of the results obtained by analytical solution and by GoldSim

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 spatial and temporal discretization on concentration profiles and diffusive outfluxes related to hollow cylindrical buffer. The aim of these calculations is to compare results of concentration profiles and diffusive fluxes from the cylindrical buffer which were calculated using the simulation tool GoldSim with the analytical solution of the non-steady state diffusion equation given in the previous section. The analytical calculations were carried out using MATLAB [5].

The calculations were performed for a stable nuclide and different distribution coefficients – 0 m^3/kg, 1 m^3/kg and 20 m^3/kg. Other transport and medium properties were assumed to be same and no solubility limit was considered in the individual calculation cases: C_1 = 1 g/l, De = 2x10^{-10} m^2/s, ε = 0.36, ρ_d = 1600 m^3/kg, a = 0.325 m, b = 0.625 m. To eliminate the discretization error, the bentonite buffer was discretized into 60, 15 and 5 cells and the concentration profiles were compared between individual cases. The concentration profiles were evaluated in the time t = 1000 yr. Apparent diffusion coefficients, calculated using a relation $D_a=D_e/(\varepsilon+\rho_d.K_d)$, are summarised in the Tab. 1.

Tab. 1. *Calculated apparent diffusion coefficients.*

	K_d = 0 m^3/kg	K_d = 1 m^3/kg	K_d = 20 m^3/kg
D_a [m^2/s]	5.56×10^{-10}	1.25×10^{-13}	6.25×10^{-15}

The concentration profiles of the nuclides with distribution coefficients 0 m^3/kg and 1 m^3/kg are shown in Fig. 2. It can be seen that the results of concentration profiles calculated using the analytical solution implemented in MATLAB and by GoldSim simulation tool (finite volume approach) are in good agreement for every spatial discretization case when we consider a fine temporal discretization (basic timestep - 1 day, medium precision setting). In cases with higher distribution coefficients (1 m^3/kg, 20 m^3/kg), the concentration in the cells is changing significantly and a very coarse temporal discretization (basic timestep – 1000 years, low precision setting) causes differences in the calculated concentrations. In the first case (when K_d is equal to 0 m^3/kg), the steady state conditions were reached and the satisfying results were obtained in every analysed case. The concentration profile can be then calculated by a simple relation $C = C_1.ln(r/b)/ln(b/a)$.

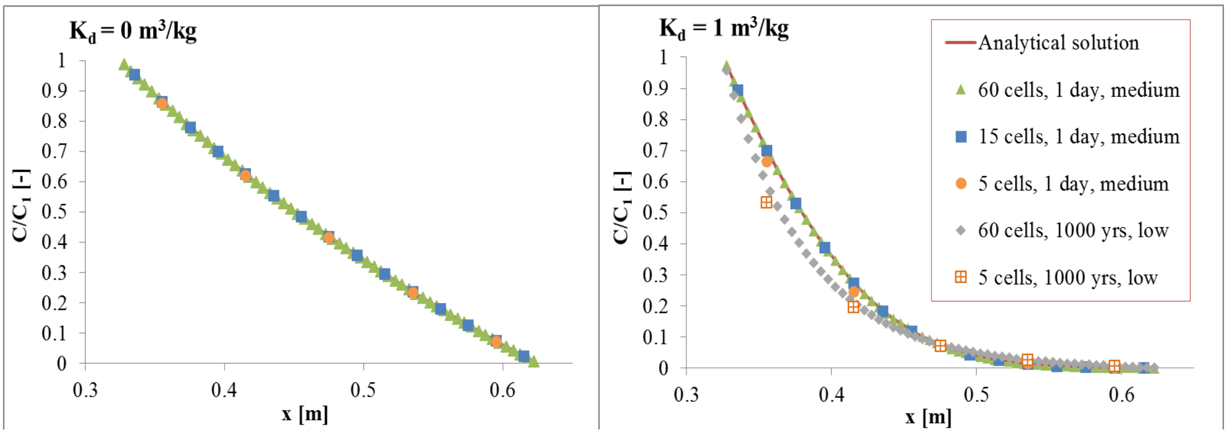

Fig. 2. *Comparison of concentration profiles of the stable nuclide.*

Also the results on diffusive outfluxes from the cylindrical bentonite buffer calculated using the simulation tool GoldSim and MATLAB (Equation 11) were compared. Given the fact that modelling of diffusion is one-dimensional, a hypothetical buffer length was determined. Due to this reason, the internal diffusive area of the modelled hollow buffer was assumed to be equal to the total outer area of the disposal container (including the top and the bottom part). Based on this assumption the hypothetical buffer lenght L = 3.995 m was determined. The diffusive outflux in terms of kg/yr was then calculated as the diffusive outflux J_t^b [kg/$m^{-2}s^{-1}$] multiplied by the outer area of the buffer ($2\pi.b.L$). As it can be seen from Fig. 3, the calculated diffusive outflux of nuclide with the low distribution coefficient is highly dependent on the timesteps specified in the simulation settings as the steady state conditions are reached very early (after approximately 5 years) and the diffusive flux is changing rapidly within the first years of the simulation. On the other hand it can be seen that the spatial discretization also influences a calculated outflux of the strongly sorbing nuclide (Fig. 3, graph on the right, closer proximity), especially when the flux is very low.

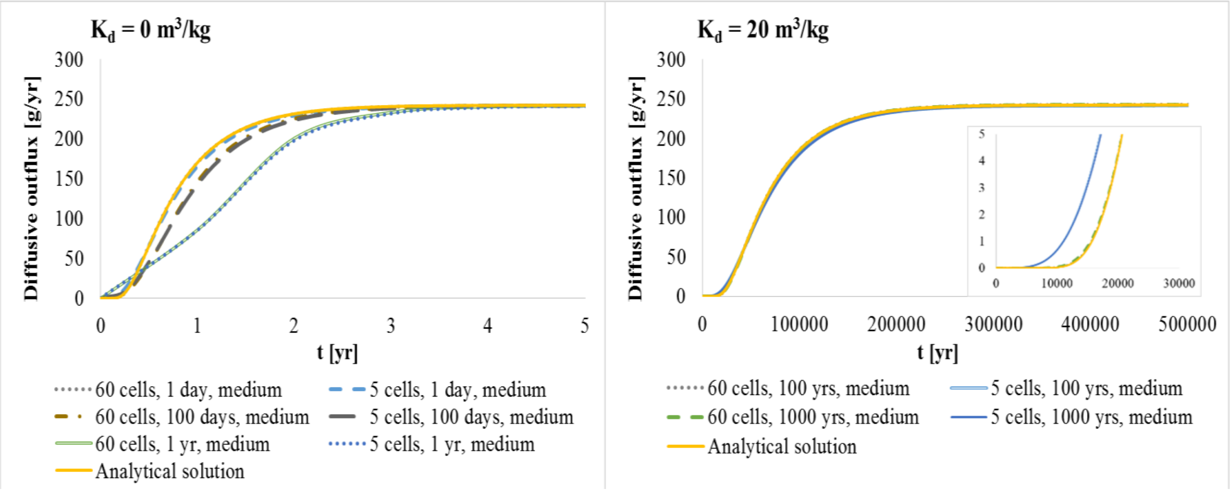

Fig. 3. *Comparison of diffusive outfluxes of the stable nuclide.*

5. Conclusion

Based on the results it can be concluded that the concentration profiles of the stable nuclide with specified different distribution coefficients calculated using MATLAB (analytical solution) and using the simulation tool GoldSim are in good agreement when the fine temporal discretization is used. In the case when $K_d = 0\ m^3/kg$, the simulation time (t = 1000 yr) was sufficient to reach the steady state conditions and the temporal as well as spatial discretization influenced the results negligibly.

The temporal discretization also significantly influences diffusive outfluxes when the diffusive mass fluxes are changing rapidly (the case of nuclides with low distribution coefficients).

However, when the accurate results on the concentration profiles and diffusive outfluxes are needed (e.g. in the case of diffusion experiments) it seems to be necessary to define not only appropriately small timesteps but also to perform an analysis of the spatial discretization to obtain as accurate results as possible.

Acknowledgement

This project has been supported by the Slovak Grant Agency for Science through grant VEGA 1/0863/17.

References:

[1] Huang, I.B., Yen, S.K. Diffusion in hollow cylinders for some boundary conditions. I. Mathematical treatment. Journal of Materials Chemistry and Physics 74, p. 289-299 (2002). [online].[cit. 25. April 2017]. <Available:<http://www.sciencedirect.com/science/article/pii/S0254058401004837 >.

[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 32, 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, Clarendon Press, Oxford (1975).

[5] The MathWorks, Inc.: MATLAB – Getting Started Guide.