Radionuclide Transport through Different Routes near a Deposition Hole for Spent Nuclear Fuel

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Abstract: Radionuclide transport modeling is a part of the research concerning geological disposal of spent nuclear fuel. Typically, the transport models near a single deposition hole focus on the reactions of nuclides, while the model geometry and the flow of groundwater are often simplified. In this paper, instead, a radionuclide transport model in a detailed 3D geometry with no reactions is introduced. The aim is to study the effects of the geometry and realistic flow fields on the nuclide transport. The groundwater flow is modeled with the continuity equation and Darcy's law, whereas the mass transport problem of a nuclide reduces to a general convection-diffusion problem. This paper focuses on development of the model instead of the final results.

Keywords: nuclear waste management, spent nuclear fuel, radionuclide transport, Darcy's law, convection-diffusion

1 Introduction

The Finnish diposal plan for spent nuclear fuel is, in brief, to seal the used fuel rods into copper-covered iron canisters, surround the canisters with bentonite (a montmorillonite clay), and place the whole packages appoximately half a kilometer deep into the bedrock. In this study, I follow a disposal concept where vertical holes are drilled in excavated, horizontal tunnels (figure 1).

Excavation of the tunnels damages the surrounding bedrock forming a so-called excavation damaged zone (EDZ). In the model, it is assumed to consist of rock with very thin water-conducting fractures, which make the EDZ a porous medium. The slow flow of water in EDZ is assumed to obey Darcy's law. Besides these induced flow paths, the bedrock includes natural fractures. These are described with three thin (aperture<1 mm), plain water conducting zones. In reality, the fractures are not necessarily smooth, straight, nor completely free of small pieces of the rock. In addition, the flow of water is slow. Thus, the momentum transport of water also in the fractures is described with Darcy's law istead of Brinkman or Navier-Stokes equations. Another water conducting part in the model is the backfilled disposal tunnel. The backfill material, however, conducts water more poorly than the EDZ or the natural fractures. The bentonite is assumed to not conduct water.Thus, radionuclides are assumed to be transported by diffusion in the bentonite and by diffusion and advection in the water-conducting part. In the model, radionuclides are released inside the canister and transported by diffusion through a small defect (5 cm deep copper-penetrating hole with 0.5 mm radius) to bentonite. The interior of the canister is not interesting in this study, hence the interior volume is assumed to be perfectly mixed and it is described with an equation on the defect boundary. In this way computational resources are saved, and possible problems caused by the tricky, small hole are avoided.

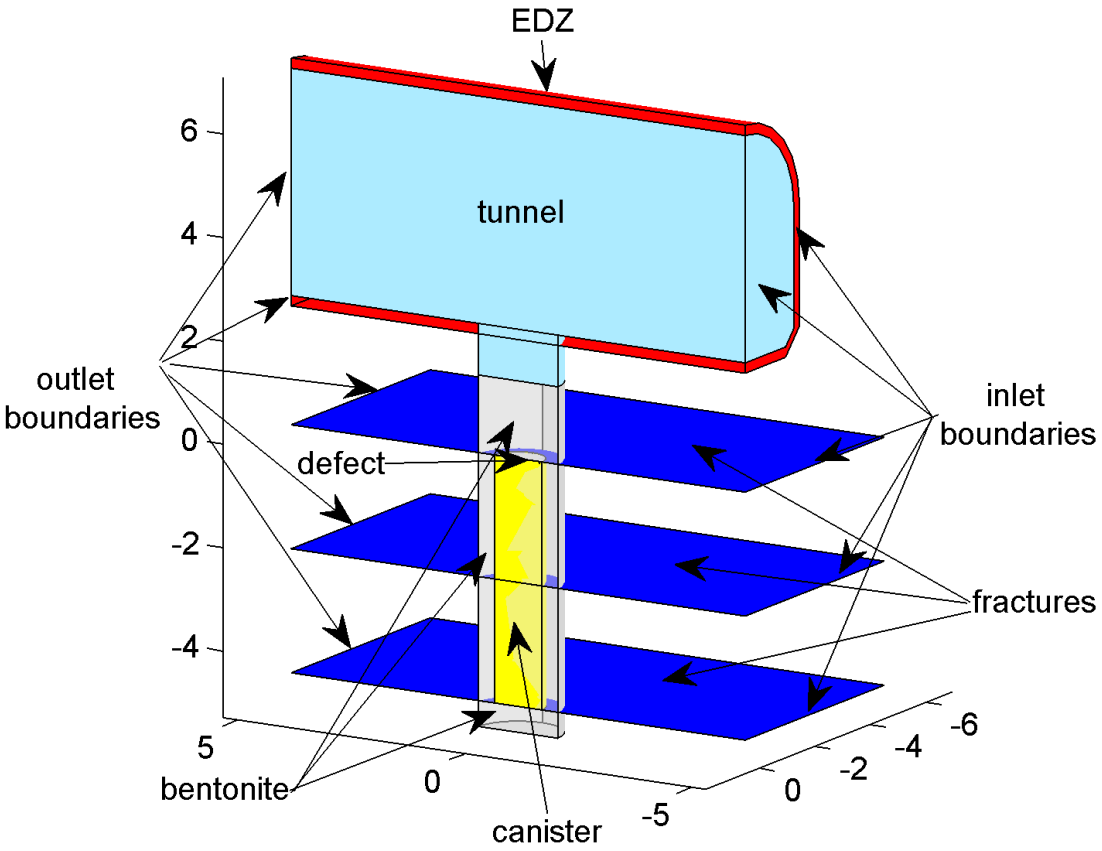

Figure 1: Geometry of the model. Computational work is reduced by halving the geometry along a symmetry plane. The scale of the axis is meters. The top part of the deposition hole is light blue since it is of the same material as the tunnel backfill. The defect in the canister is so small that it is hard to see in the figure. The defect is placed on the symmetry plane near the right edge of the canister.

2 Governing Equations

Darcy's law is

$$\boldsymbol{U} = -\frac{\boldsymbol{\kappa}}{\mu}(\nabla p - \rho \boldsymbol{g}),$$

where $\boldsymbol{U}$ is the Darcy velocity, $\boldsymbol{\kappa}$ the permeability tensor, μ the viscosity of water, p the pressure, ρ the density of water, and $\boldsymbol{g}$ the gravitational acceleration. Porous media flow obeys this law when the flow velocity and the shearing forces of the fluid are low. Combining the law with the time-independent continuity equation for an incompressible fluid in a porous medium, we have an equation for the flow of water. The water flow problem is to find the pressure $p = p(\boldsymbol{x})$ such that

$$\begin{cases} \nabla \cdot [-\frac{\boldsymbol{\kappa}}{\mu}(\nabla p - \rho \boldsymbol{g})] = 0 & \text{in } \Omega_{flow} \\ p = p_{in} & \text{on } \partial\Omega_{inlet} \\ p = p_{out} & \text{on } \partial\Omega_{outlet} \\ \boldsymbol{n} \cdot \left(-\frac{\boldsymbol{\kappa}}{\mu}\nabla p\right) = 0 & \text{elsewhere on } \partial\Omega. \end{cases}$$

In the study, the water-conducting zones are assumed isotropic. Thus, the permeability tensor can be substituted with a scalar.

Having the pressure, the Darcy velocity $\boldsymbol{U}$ can be computed from Darcy's law. It is needed in the mass transport problem, which reads: find the radionuclide concentration $c =$

$c(\boldsymbol{x}, t)$ such that

$$\begin{cases} \theta \dfrac{\partial c}{\partial t} + \nabla \cdot (-D_e \nabla c) + \boldsymbol{U} \cdot \nabla c = 0 \\ \qquad \text{in } \Omega_{flow} \\ \theta \dfrac{\partial c}{\partial t} + \nabla \cdot (-D_e \nabla c) = 0 \\ \qquad \text{in } \Omega_{bentonite} \\ -\boldsymbol{n} \cdot (-D_e \nabla c) = \dfrac{D_e}{l} (c_c - c) \\ \qquad \text{on } \partial\Omega_{defect} \\ \boldsymbol{n} \cdot (-D_e \nabla c) = 0 \qquad \text{on } \partial\Omega_{outlet} \\ \boldsymbol{n} \cdot (-D_e \nabla c + c\boldsymbol{u}) = 0 \qquad \text{on } \partial\Omega_{elsewhere} \\ c(\boldsymbol{x}, 0) = 0 \qquad \text{in } \Omega, \end{cases}$$

where θ is the porosity, D_e the effective diffusivity, $\boldsymbol{u}$ the Darcy velocity, $\boldsymbol{n}$ the surface normal vector, l the defect depth, and c_c the canister interior concentration. c_c is solved simultaneously with the above equation from the following initial value problem:

$$\begin{cases} \theta \dfrac{\partial c_c}{\partial t} = -\dfrac{D_e A}{Vl} (c_c - c) \\ c_c(0) = c_{c,0} \\ \theta \dfrac{\partial c_c}{\partial t}(0) = -\dfrac{D_e A}{Vl} (c_c(0) - c(\boldsymbol{x}, 0)), \end{cases}$$

where $\boldsymbol{x} \in \partial\Omega_{defect}$, A is the defect cross-section area, V the canister interior volume, $c_{c,0}$ the canister interior initial concentration, and $\partial c/\partial t(0)$ is the initial condition for the interior concentration time derivative. This initial value problem is a model of a perfectly mixed volume, from which a solute is transported by diffusion via a small hole defined by the parameters above.

3 Methods

The used elements are various orders of Lagrange elements and third order Hermite elements [1][2]. The numerical stabilization tests have been done with second order Lagrange elements, whereas the comparison of elements includes second and third order Lagrange elements and third order Hermite elements.

Linear equation systems are solved with direct solvers, mainly with Umfpack solver but Pardiso solver is also used in some occasions. The time-dependent differential algebraic equation (DAE) system (or ODY system, if no Dirichlet boundaries exist) produced by the finite element discretization is solved with the Backward Differentiation Formula (BDF) solver. In most test cases, the maximum BDF order is five, but in some cases the maximum order is restricted to two, which makes the method A-stable for linear systems. The Generalized-α method may have some beneficial oscillation damping properties, but it has not been tested yet.

3.1 Numerical stabilization

The convection-diffusion problem for the mass transport needs stabilization to damp the observed spurious oscillations, which derive from the geometric scale differencies and from the parameter differencies between the materials. These scale and material differencies cause steep concentration gradients, which again are numerically problematic and cause oscillations. In the study, the Streamline Upwind/Petrov-Galerkin (SUPG) stabilization is used with different stabilization parameters to damp these oscillations. Also, shock-capturing technique [4] is tried to damp the oscillations in the cross-wind directions.

A general convection-diffusion problem reads: find $u = u(\boldsymbol{x}, t)$ such that

$$\begin{cases} \dot{u} + \boldsymbol{\beta} \cdot \nabla u - \varepsilon \Delta u = f & \text{in } \Omega \\ u(\boldsymbol{x}, t) = u_b(t) & \text{on } \Gamma_D \\ \varepsilon \nabla u \cdot \boldsymbol{n} = g & \text{on } \Gamma_N. \end{cases}$$

In the model, $\dot{u} = \partial c/\partial t$, $u = c$, $\boldsymbol{\beta} = \boldsymbol{U}/\theta$, $\varepsilon = D_e/\theta$, and $f = 0$. With residual

$$R = \dot{u}_h + \boldsymbol{\beta} \cdot \nabla u_h - \varepsilon \, \Delta u_h - f$$

a consistently stabilized finite element formulation of the problem is: find $u_h = u_h(\boldsymbol{x}, t) \in U_h = \{u_h | u_h \in P_n \text{ and } u_h = u_b \text{ on } \Gamma_D\}$ such that

$$B(u_h, v) + \sum_K \int_K R \, \tau_s \, P \, d\boldsymbol{x} = l(v) \quad \forall v \in V_h$$

where

$$\begin{aligned} B(u, v) &= \int_\Omega \dot{u} \, v \, d\boldsymbol{x} + \int_\Omega (\boldsymbol{\beta} \cdot \nabla u) \, v \, d\boldsymbol{x} \\ &\quad + \int_\Omega \varepsilon \, \nabla u \cdot \nabla v \, d\boldsymbol{x}, \\ l(v) &= \int_\Omega f \, v \, d\boldsymbol{x} + \int_{\Gamma_N} g v \, ds, \end{aligned}$$

U_h and $V_h = \{v | v \in P_n \text{ and } v = 0 \text{ on } \Gamma_D\}$ are the finite element spaces, K an element, τ_s

the stabilization paremeter, and P the stabilization test function. In the SUPG stabilization $P = \boldsymbol{\beta} \cdot \nabla v$. In most computations, the used stabilization paremeter is constant. The problem with such stabilization is that in some parts of the model the stabilization might not be strong enough and in other parts it may be excessive decreasing accurary. Thus, a stabilization parameter suggested in [3] would be preferable:

$$\tau_s = \frac{h}{2|\boldsymbol{\beta}|p}\xi(P_e^p),$$
$$\xi(P_e^p) = \max\{0, 1 - 1/P_e^p\} \qquad \text{and}$$
$$P_e^p = \frac{|\boldsymbol{\beta}|h}{2\varepsilon p}.$$

Here h is the element size, P_e^p the local, approximation degree dependent Peclet number, and p the approximation degree, that is, the element order. This τ_s should provide reasonable amount of stabilization in all model regions, but the use of it has still been unsuccesful due to some convergence difficulties. One possibility to solve these problems is to limit this stabilization parameter from above to avoid very large parameter values. This can be done with

$$\tau_{s,L} = \min\{L, \tau_s\}$$

where L is the limit. The results of this approach are not yet ready.

Whereas the SUPG stabilization works in the streamline direction, the shock-capturing technique stabilizes the solution in the crosswind direction. It is needed when the changes in concentration are steep not only in the gradient direction but also in other directions. An important feature of such stabilization is that the consistent version makes the problem nonlinear even if the initial problem is linear. The technique has been tested with constant tuning parameter. The benefits seems to be litte in comparison to the increase in computation time. More sophisticated shock-capturing stabilization parameter choices can be found in literature, e.g. [4] or [3], but they have not been used.

3.2 Weak Form of Boundary Problem

The boundary equation describing the canister interior has to be formulated in weak form for COMSOL. The weak form is: find c_c on the defect boundary such that

$$\int_{\partial\Omega_{defect}} \theta \dot{c_c}\, c_{c,test}\, d\boldsymbol{x} + \int_{\partial\Omega_{defect}} -\frac{D_e A}{Vl}\,(c_c - c)\, c_{c,test}\, d\boldsymbol{x} = 0$$

and c_c satisfies the initial and boundary conditions. The left-hand side integrand is the **dweak** term in COMSOL and the right-hand side integrand the **weak** term.

4 Use of COMSOL Multiphysics

The Chemical Engineering Module is used for both the water flow and the mass transport problems. Stabilization techniques are included in the module, thus it is the obvious choice for the mass transport problem. The porous media equations have to be systematically modified to fit into the module. The module also includes a momentum transport option based on Darcy's law, hence the water flow problem can be easily solved with the same module.

The equation on the defect surface is solved with Boundary Weak Form application mode in PDE Modes in COMSOL Multiphysics main package. The equation solved has to be written in the weak form, but that is straightforward as seen in the section above.

The model has been scripted in Matlab with the help of the graphical user interface. Firstly, a script that initializes the model geometry was written. Then the model geometry was opened with the COMSOL graphical user interface (GUI), with which the physics were defined. The automatically produced m-file was saved. The mesh was created again by scripting, whereas the solution algorithms were set with the GUI. The long m-file obtained was divided into convenient parts and a master m-file was written. The scripting procedure requires some work, but the result is a set of m-files, which can be used to vary the model parameters easily.

The main benefits of the Matlab scripting are the automated mesh generation, the ability to change the geometry in a flexible manner, and the overall sequencing capabilities. In the model, prism mesh elements are used in the fractures, hence the mesh has to be created in a strict order. The scripted mesh generation

helps substantially, if the mesh density has to be changed in any way. The changes in geometry are usually laborious, but they can be done quickly with scripted geometry generation described above. The sequencing capability is valuable when a reasonable combination of mesh, element, and solver paremeters is sought. Also, a scripted model-initializing sequence is useful, if the modeled problem has to be solved with varying physical paremeters or with different boundary conditions.

5 Experimental Results

An example of the results is shown in figure 2. The flowing water transports the radionuclide out from the model before the nuclide has time to diffuse far in the flowing parts of the model.

Negative concentrations in some regions of the model often indicate problems in the numerical solution. Thus, the negative concentration is used to demonstrate the effects of different stabilization techniques on the solution in the following figures 3–5. The solution with no stabilization oscillates visibly. SUPG stabilization damps the most of the oscillation, but shock-capturing technique seem to have little effect on the remaining oscillations. Surprisingly, the SUPG stabilization also reduces the computation time compared to the solution with no stabilization.

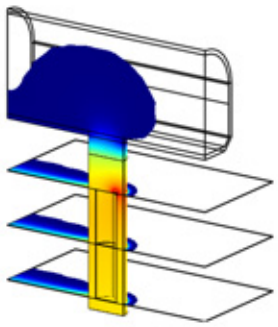

Figure 2: An example of the concentration profile at $2.4{\cdot}10^5$ years. The scale is from $1{\cdot}10^{-8}$ (blue) to $2{\cdot}10^{-5}$ (red) grams per liter. The SUPG stabilization was used with constant tuning parameter.

The stabilization seems to help the time-dependent solver to take larger time-steps than without the stabilization. The SUPG with the shock-capturing technique requires somewhat more computation time than only SUPG (95 min versus 65 min), but the solution changes only a little.

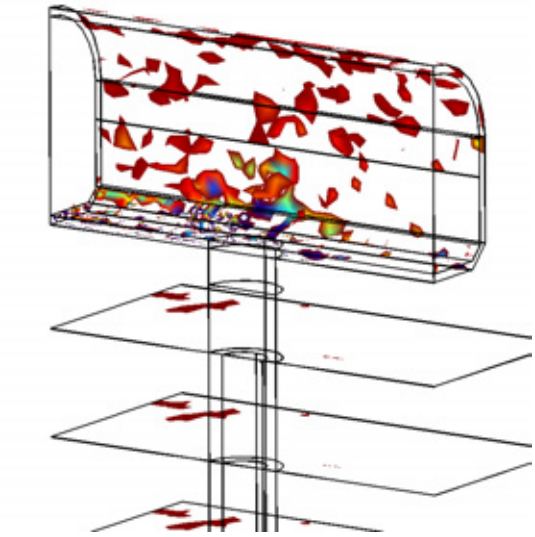

Figure 3: The regions with negative concentration in the solution with no stabilization at $2.4{\cdot}10^5$ years. The computation time was 73 minutes.

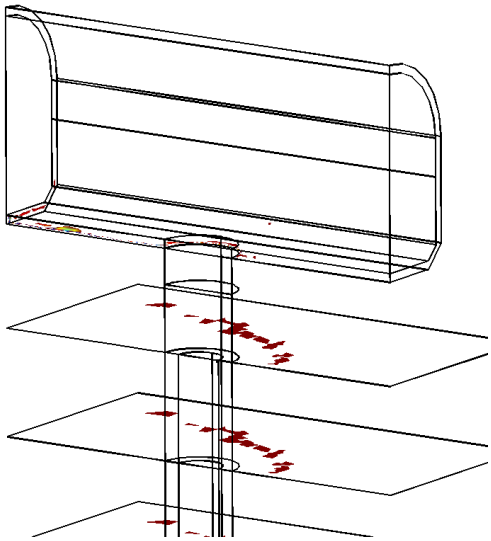

Figure 4: The egions with negative concentration in the solution with SUPG stabilization at $2.4{\cdot}10^5$ years. The stabilization paremeter is constant. The computation time was 65 minutes.

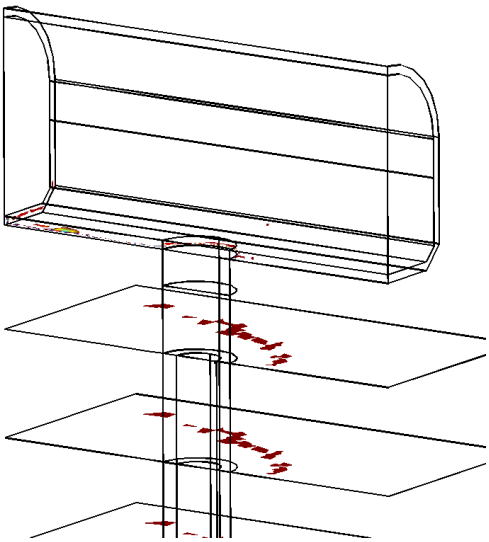

Figure 5: The regions with negative concentration in the solution with SUPG and shock-capturing stabilization at $2.4 \cdot 10^5$ years. The stabilization paremeters are constants. The computation time was 95 minutes.

A problem with finite element methods is that they do not necessary preserve mass locally. The Hermite element has the gradients at the mesh element nodes as degrees of freedom. Thus, they could be thought to have better local mass balance properties.

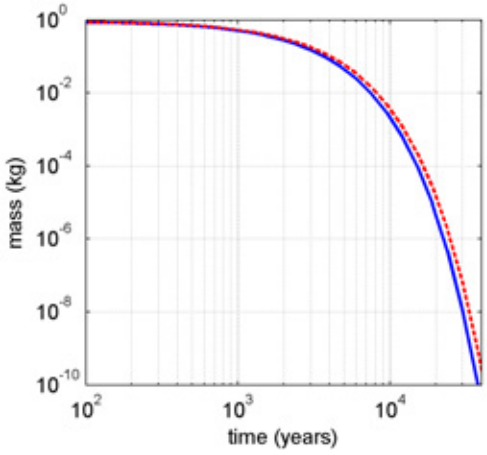

Figure 6: A comparison of the elements. The total mass in the interior of the canister and in the bentonite as a function of the time. The figure is from a test case where there is only one fracture and no tunnel section in the model. The blue line is the result with third order Hermite elements and the red slashed line is the result with second order Lagrange elements.

The minimum order of the Hermite element is, however, three making the computation time long if dense mesh is used. In addition, high order elements might not be an ideal choice for a model with steep gradients because of their tendency to make the solution oscillate. If time behaviour of the total mass in the bentonite and inside the canister is studied, local mass balance is not critical since the region of interest is quite large. Of course, the solution has to be otherwise reasonable. Figure 6 shows that the element choice affects the solution slightly.

6 Discussion

COMSOL Multiphysics is a flexible tool when studying non-reactive transport of one radionuclide in the spent nuclear fuel final deposition environmnent. The realistic geometry generation possibilities and the control of the solution procedure are valuable properties. The number of the nuclides could be easily increased, radioactive decays added within certain limits, and sorption according to the linear sorption law used. The limiting factor is the computation resources. Also mechanical dispersion could be easily used if wanted. It, in fact, stabilizes the problem a bit.

Problems arise when the solubilities of the nuclides are limited. This is especially problematic when multiple nuclides of the same element share the solubility. The solubility handling could be built into COMSOL, but it would require a significant amount of studying and testing to make it work properly. It seems that, other application specific software are needed for models including the above chemical phenomenon. On the contrary, COMSOL often handles transport phenomena better than the specific software.

A good feature of COMSOL is its applicability to numerous problems. The techniques used in the study in this paper can be extended to other problems of similar type, like in [5]. The techniques, however, may need some modifications.

7 Conclusions

Non-reactive radionuclide transport problems in realistic geometry can be solved effectively with COMSOL Multiphysics. If, however,

some special chemical phenomena are considered, application specific software is needed. The techniques used in the model in this paper are applicable to other problems of similar type.

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