

Thermo-Hydro-Mechanical-Chemical (THMC) Modelling of the Bentonite Barriers in Final Disposal of High Level Nuclear Waste

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Abstract: The bentonite barrier is an essential part of safe nuclear waste repository in granitic bedrock. In this work COMSOL Multiphysics® is applied to THMC modelling meaning full coupling of thermal (T), hydrological (H), mechanical (M) and chemical (C) phenomena and processes taking place in a bentonite buffer. The system is studied in different geometries, which consists of an overall 3D layout and a 2D cross-section of bentonite buffer and open fracture. Altogether three main periods have identified in the modelling: 1) emplacement of canister, 2) saturation, and 3) long term safety. The main interest has been on diffusional transport of different species into and out of bentonite buffer.

Keywords: nuclear waste repository, bentonite, transport, THMC.

1. Introduction

1.1 Nuclear waste management

The basic option in Finnish nuclear waste management is geological disposal for all types

of nuclear wastes: As for the financing, Finland follows the ‘polluter pays’ principle meaning that nuclear energy generating power companies have full responsibility to plan, finance and implement the safe management of their nuclear waste. The power companies in Finland, Fortum and Teollisuuden Voima, founded jointly the nuclear waste company Posiva Oy in 1995 for the disposal of their spent fuel.

Repositories excavated in bedrock for low- and intermediate level waste are in operation at nuclear power plant sites: one in Eurajoki, and one in Loviisa. Decommissioning waste are planned to be disposed of in extensions to these repositories.

Extensive preparations are underway for spent nuclear fuel disposal in one national repository at Olkiluoto in Eurajoki, see the KBS-3 disposal concept in **Fig. 1**. The next milestone in Finnish spent fuel management will be the Construction Licence application for the underground disposal facility at Olkiluoto that Posiva is planning to submit to the Finnish Government in 2012.

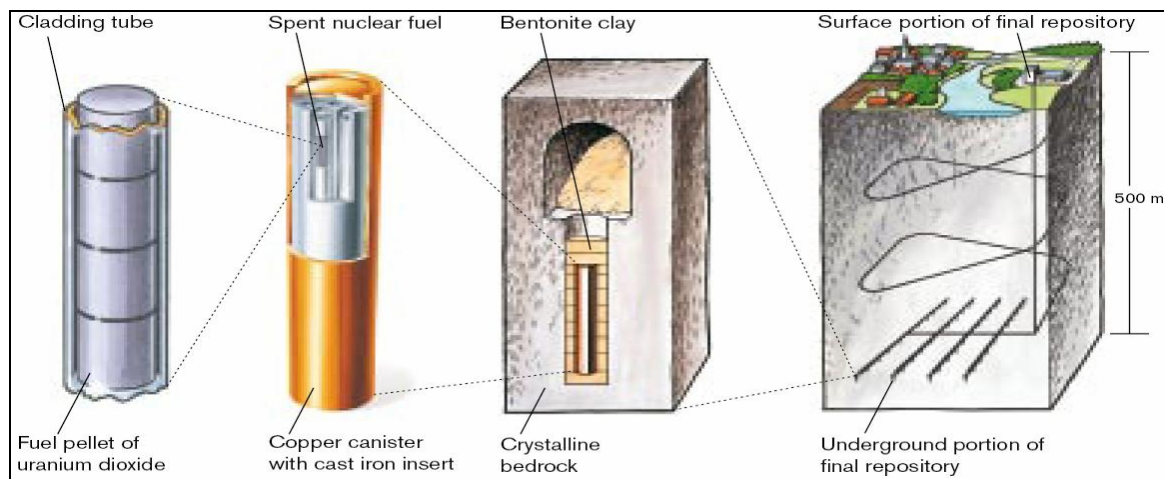


Fig. 1. The KBS-3 disposal concept for spent nuclear fuel of nuclear wastes from [SKB, 2006]

1.2 Modelling problems

Many phenomena and processes have to be understood when considering the safety of engineering barrier system (EBS) of the spent fuel repository **Fig. 1**. For this understanding important tool is modelling which is done mainly computationally. In this work the main interest is in the bentonite buffer part of the EBS. Its main functions in terms of different process classes (see **Fig. 2**) are

- o to minimize hydraulic conductivity near waste canisters (hydraulic processes (H)),
- o to maintain beneficial chemical environment (chemical (C) and thermal (T) processes),
- o to delay or limit release of radionuclides (CH) and
- o to limit the stresses acting on the canister (mechanical processes (M)).

The chemical phenomena taking place in a bentonite barrier in a repository depend directly on temperature, the composition of interacting fluid, the diffusion coefficients of solutes, the reactive surface area of minerals, the texture of porous media, the nature and amount of accessory mineral, radioactive decay of nuclides etc. Thermal gradient from the highly radioactive spent fuel, affects chemical phenomena and diffusion, advection and convection (THC). Adding the swelling (M), the alteration of bentonite and the copper canister the modelling becomes quite challenging. Typically, models including mechanical behaviour handle chemical

reactions in a quite simplified way and vice versa.

It is quite hard to consider all phenomena in one model, because of their complexity and mutual couplings. Anyhow, there are many coupled models, which try to describe the evolution of bentonite buffer as realistically as possible, but because of great complexity of phenomena and processes they are all somehow simplified. Nowadays, the computers capacity has increased so much that the researchers are able to move from simple one dimensional methods to more complex ones including coupled modelling (2D, 3D) where for example chemical reactions and hydrological behaviour are considered (C-H).

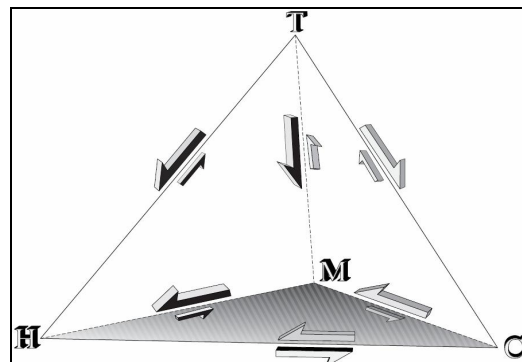


Fig. 2. Schematic view of the couplings between the thermal (T), hydrological (H), mechanical (M) and chemical (C) processes. The estimated relative importance of a coupling is indicated by the thickness of the arrow, from [Rasilainen et al. 1999].

BUFFER

- plastically isolates the canister from rock and protects it against minor rock displacements
- keeps the canister in place in the deposition hole
- mass transport predominantly by diffusion
- conducts the heat from canister to the rock
- has sufficient permeability to gases
- filters colloids and prevents growth of microbes
- chemically and mechanically stable
- no harmful effects on other barriers

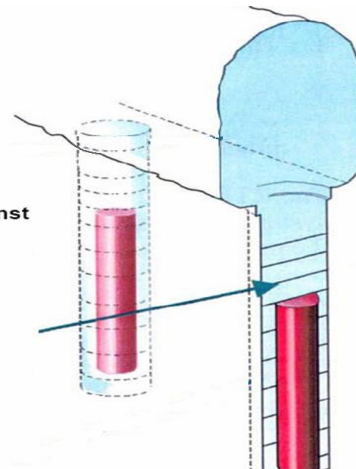


Fig. 3. The functions of bentonite buffer, the tunnel backfill and sealing structures. Modified from [Pastina & Hellä 2008]

1.3 How to solve the problems?

VTT has a versatile modelling toolbox for the whole chain of analyses required in the safety assessment of nuclear waste disposal. There are detailed research models as well as more simplified safety assessment models. Having several parallel models available for the solving of problems reduces conceptual and numerical uncertainty.

A brief description of some codes at VTT for the bentonite modelling in the near-field is given below. As indicated in **Fig. 3** we consider near-field to include various coupled processes (e.g. THC, THM) the modelling of which requires coupled models. Each code mentioned below has a specific area of application based on its particular properties (pros and cons). COMSOL Multiphysics® is the main tool for the current study and it will be described more detailed way than the others.

EQ3/6

EQ3/6 [Wolery & Jarek, 2003] is a software package utilized to perform geochemical modelling calculations including fluid-mineral interaction and solution-mineral equilibrium in aqueous systems. The software includes thermodynamic equilibrium, irreversible mass transfer and reaction kinetics. There are two major components: EQ3NR is a speciation-solubility code and EQ6 is a reaction path modelling code to simulate irreversible water/rock interaction and/or fluid mixing with kinetics. It has its own thermodynamic editable databases which allow the use of different chemical activity models. It is a useful tool for modelling the chemistry in final repository but it is only “zero”-dimensional code without mechanics and migration.

PetraSim

PetraSim [Thunderhead engineering, 2005] is a user interface which is a interactive pre/post-processor for TOUGH2, T2VOC, TMVOC, TOUGHREACT, TOUGH-Fx/HYDRATE and TETRAD simulators. PetraSim helps users to develop models and view result of models based on (TOUGH2) nonisothermal flows of multicomponent/-phase fluids in one, two, and three-dimensional porous and fractured media.

T2VOC and TMVOC include three-phase flows of water, air and volatile organic chemicals. The TOUGHREACT adds chemical reactions to PetraSim and the TOUGH-Fx/HYDRATE the capability to represent methane hydrates. PetraSim is also a capable tool for simulating the final repository but the code has also many problems and the user manual is concise. In addition it lacks the tools to construct realistic and modern geometries.

GoldSim

GoldSim [GoldSim Technology Group LLC, 1998-2007] is a system management program which has a module for simulating radionuclide transport. The program can be used to manage the whole deposition environment by linking necessary programs to it. However, at VTT it has so far mainly been used to simulate transport phenomena. Nuclides behave independently in radioactive decay and as a part on an element in chemical reactions making computing challenging. GoldSim can handle this but only in simple geometries with limited transport phenomena.

1.4 Why and what by COMSOL?

COMSOL MultiPhysics is a very flexible toolbox for solving problems including partial differential equations. It enables full coupling between physical quantities and their governing equations. The use of COMSOL is easy at starting level, but on the other hand very complicated couplings require elaborate programming.

For nuclear waste management the Earth Science module appears most suitable of COMSOL's application packages. While, it includes many interesting phenomena and processes, like fluid flow and solute transport both in fully and partially saturated porous medium, there are difficulties in chemical modelling compared to, e.g., EQ3/6 and PetraSim. On the other hand, quite complicated, realistic, geometries can be modelled by COMSOL and even the meshing is becoming easier.

Therefore, COMSOL suits best for modelling overall transport in bentonite buffer, groundwater and rock fracture system. It is even possible to model transport from thin fracture to bentonite

buffer in a realistic way. COMSOL includes also heat transport and it is possible to couple hydraulic and thermal modelling to mechanistic calculation.

2. Governing Equations

Both granitic rocks and bentonite are porous and in some cases not fully saturated by water in application of nuclear waste management. Therefore, instead of Navier-Stokes equation for flow velocity and Fick's laws for diffusional transport, special versions of these equations are used. The flow velocity follows Darcy's law in case of full saturation

$$S \frac{\partial p}{\partial t} = -\nabla \cdot \mathbf{u} + Q_s = \nabla \cdot \left[\frac{\kappa}{\eta} (\nabla p + \rho \mathbf{g}) \right] + Q_s \quad (1)$$

where S is the storage coefficient, p is the pressure of fluid, \mathbf{u} is the flow velocity, Q is a fluid source, κ is the permeability of the porous medium, η is the dynamic viscosity of the fluid, ρ is the density of the fluid, \mathbf{g} is the gravitational acceleration. In variable saturated porous medium a more complicated Richard's equation should be applied.

In Fick's laws for porous media, instead of molecular diffusion constant, an apparent diffusion constant

$$D_a = \frac{G\phi}{\alpha} D_w \quad (2)$$

is applied. G is a geometric factor to diffusion, ϕ is porosity of the medium, α is the capacity factor and D_w is the diffusion coefficient in the fluid. Equation for solute transport in fully saturated case is written as

$$\alpha_i \frac{\partial c_i}{\partial t} = -\nabla \cdot \mathbf{J}_i + R_i = \nabla \cdot [D_a \nabla c_i - \mathbf{u} c_i] + R_i \quad (3)$$

where α_i is the capacity factor, c_i is concentration, \mathbf{J}_i is the diffusive flux, R_i is a source term, \mathbf{u} is the velocity of the fluid and i denotes species.

Chemical reaction may be kinetically controlled (included in Eq. (3)) or equilibrium controlled (modelled by mass action and mass balance). Capacity factor, α , includes surface reactions, which can be theoretically modelled

by surface complexation and ion exchange reactions, instead of empirical α -values.

At disposal depth, water flows mainly in very thin (aperture 1 mm or below) fractures and on the average very slowly. Diffusion constants vary from free water diffusivity values (about $3 \cdot 10^{-9} \text{ m}^2/\text{s}$) to several orders of magnitude lower values.

Transport of heat is modelled by equation parallel to Eq. (3), while mechanical stresses acting in buffer are modelled by the basic equations of continuum mechanics.

3. Systems to be studied

At least the following systems appear interesting in studies of buffer behaviour

1. 3D model over the whole buffer volume and surrounding rock, including various fractures cutting the deposition whole; scale $10 \times 10 \times 8 \text{ m}^3$ (see **Fig. 4**)
2. 3D model around the bentonite buffer and fracture cutting it, either cylindrical or a bit simplified rectangular geometry; scale $2 \times 1 \times 0.5 \text{ m}^3$ (see **Fig. 5**)
3. 2D model for diffusion or heat transport calculations between fracture and buffer, combined possible with chemical reactions; scale $0.5 \times 0.5 \text{ m}^2$ (see **Fig. 6**)
4. 1D model for transport between the fracture and buffer in cases of very complicated chemical reaction combined with variable saturated and swelling buffer bentonite.

From disposal point of view some relevant time scales are

- 1) emplacement of canister: from few days to months
- 2) saturation of bentonite by water, from months to hundreds of years
- 3) long term safety: from hundreds of years to many thousand years, especially post-glacial periods

In all cases where the very thin fracture is included, applying numerical methods and especially meshing are quite demanding.

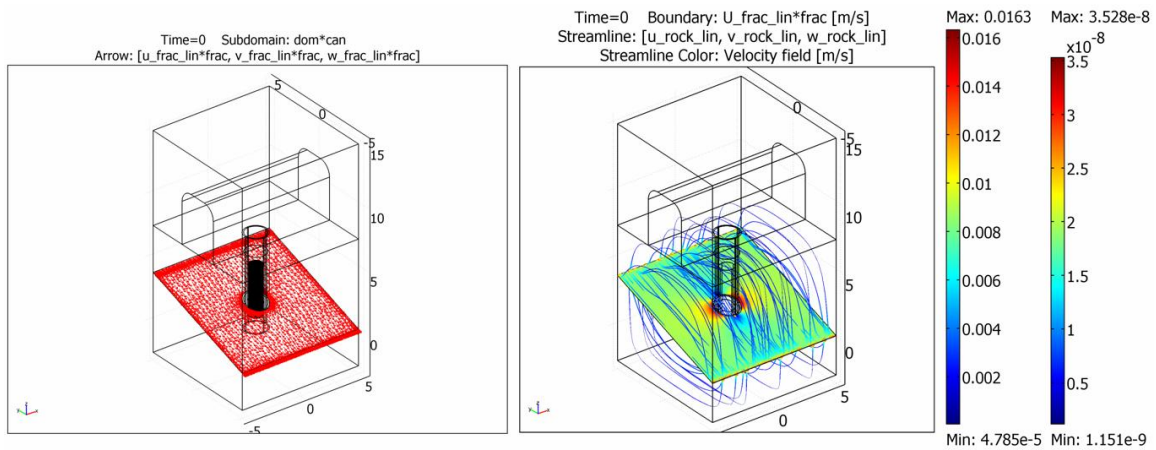


Fig. 4. 3D model of the disposal site. The velocities are computed with Darcy's law. The velocity field is represented by arrows in the left figure and by streamlines in the right figure. The dimensions of the system are:

- copper canister: radius = 0.53 m, height = 4.8 m
- bentonite: thickness = 0.35 m, height = 1+4.8+1 m
- rock block: 10m x 10m x 8m. The tunnel section is not yet in use, but gives a rough idea of the scale.

4. Models created

Most of our COMSOL-models include only transport by diffusion and groundwater flow. In the cases of this paper diffusion coefficient is set to value 10^{-10} m²/s in buffer and in fracture, and permeability in fracture is 10^{-9} m². Diffusion in rock matrix and permeability in bentonite buffer are considered to be negligibly low.

None of our present COMSOL-models includes any detailed chemical reactions, and only some transport of heat.

For most demanding calculations a Dell Precision T5400 workstation with two quad core processors and 32 GB of memory was applied.

5. Preliminary Results

This far our applications of COMSOL have been, mainly testing its capabilities in the field of nuclear waste management. Therefore, the sample of results, shown in **Figs. 4-6**, represents only test cases, except that geometries applied are realistic.

It has been considered very important to study the effect of fracture aperture, placement and direction with respect to deposition whole. By starting from simple transport models in realistic 3D geometry it is possible to simplify in a proper way the geometry to allow inclusion of complicated chemical reactions.

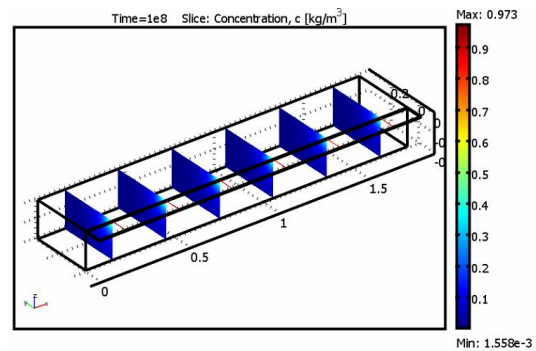


Fig. 5. Simplest possible 3D-geometry: bentonite around the canister is "straightened" to a rectangular slab and the fracture is front of bentonite. Water flows along fracture and diffusion is going both in horizontal (towards canister) and vertical directions

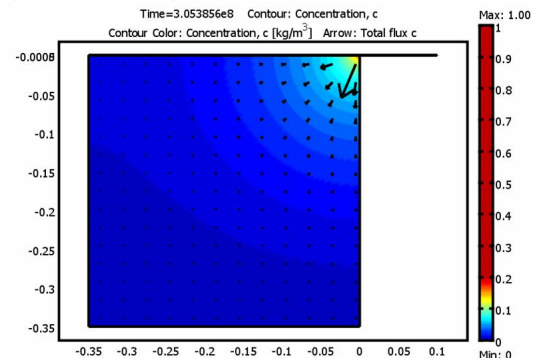


Fig. 6. Simple 2D-geometry, which enables the use of very thin fracture, but lacks groundwater flow. Diffusion in two directions: horizontally, where on the left side of the figure is the canister, and vertically, where there are possibly several meters of bentonite.

6. Discussion

The main challenge in our THMC modelling problem is to couple the various phenomena that are challenging even alone. To be able to compute results and to interpret them, some compromises have to be done. Chemical reactions can be calculated precisely, but then the geometry and transport phenomena have to be simple or vice versa. Getting the best of the both extremes requires multiple models which are simplified from the real 3D geometry step by step. In nuclear waste applications several models of each step may be required.

Besides couplings, geometry causes some problems. The scale difference between the thin 1 mm fracture and the bentonite buffer is huge. Because of this, extra care must be taken when applying numerical methods. Not surprisingly, meshing becomes difficult in some cases. To make computing efficient, in some models the fracture has been considered as a two dimensional object. This speeds up the computing significantly and works for Darcy's law, but coupling of the phenomena becomes difficult. However, meshing tools in COMSOL beat the corresponding tools in many other programs.

In this work we have observed some room for improvements in COMSOL in nuclear waste applications. Hopefully, for the future calculations, chemical reaction both in fluid and surface phases will be easier to couple to other modelling. Even in Chemical Reaction Engineering Lab® (CREL), especially equilibrium chemistry, surface reactions and mineral dissolution/precipitation appears rather clumsy to model in the present version of COMSOL.

The performance of the workstation used for demanding calculations was satisfactory. The 32 GB of memory is enough for our models, but the gain of using two quad cores has not been as large as expected.

7. Conclusions

COMSOL appears to suit quite well for transport studies even in our most complicated geometries, although we have tested it only in EBS cases. It is a good tool for testing the effects of meshing due to its relatively efficient and simple meshing tools compared, for example to

PetraSim. On the other hand, inclusion of chemical reactions in equilibrium is very clumsy (forward and backward kinetics) or must be done by Comsol Reaction Engineering Laboratory (CREL). It looks that also CREL is in difficulties with heterogeneous mineral reactions.

8. References

1. GoldSim Technology Group LLC 1998-2007. GoldSim User's Guide Version 9.60.
2. Pastina, B. & Hellä, P. 2008. Expected Evolution of a Spent Nuclear Fuel Repository at Olkiluoto (revised). Posiva Report 2006-05.
3. Rasilainen, K., Luukkonen, A., Niemi, A., Olin, M. & Pöllä J. 1999. The feasibility of modelling coupled processes in safety analysis of spent nuclear fuel disposal. Espoo: VTT - Technical Research Centre of Finland.
4. SKB 2006. Long-term safety for KBS-3 repositories at Forsmark and Laxemar - a first evaluation. Main report of the SR-Can project. Swedish Nuclear Fuel and waste Management Co (SKB);Stockholm, Sweden. SKB Technical Report TR-06-09
5. Thunderhead engineering 2005. PetraSim User Manual.
6. Wolery, T. W. & Jarek, R. L. 2003. SOFTWARE USER'S MANUAL EQ3/6, Version 8.0. Sandia National Laboratories.

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