Benchmarking as Key Element of Confidence Building in Safety Assessments for Radioactive Waste Disposal

E. Weetjens
SCK•CEN (the Belgian Nuclear Research Centre)
Boeretang 200, BE-2400 Mol; eef.weetjens@sckcen.be

Abstract: In safety cases for disposal of radioactive waste, currently in development in several countries worldwide, verification of the codes applied for assessment of the long-term repository safety is typically one of the quality assurance requirements demanded by regulatory bodies. Code verification can be done by performing benchmark calculations between two numerical codes. In this paper, an internationally organized benchmark exercise is presented, in which the intercomparison was carried out for a selection of key radionuclides, among which an actinide chain. Besides a strict comparison of the calculated output, both applied codes were evaluated on the feasibility of implementation of processes, especially focusing on realism in the source term formulation. The examples presented in this paper show that COMSOL Multiphysics can be a valuable complementary modelling tool, adequately representing most of the processes of importance in a radioactive waste repository, for varying and even extreme timescales.

Keywords: Radioactive waste disposal, vitrified high-level radioactive waste, safety assessment, code verification, benchmark.

1. Introduction

In all nuclear power generating countries, management of spent nuclear fuel and long-lived radioactive waste is an important environmental issue today. Disposal in deep geological formations, such as clay layers, is one of the promising options to dispose of these wastes. The safety assessment is an essential element of demonstrating that geological disposal is a safe and feasible long-term solution. It generally consists of an integrated series of models representing the processes of importance that govern the release and transport (both extremely slow processes) of radionuclides from the repository through various geological media up to the biosphere, where the radiological impact is usually expressed in terms of dose, for comparison with radiological protection criteria.

In a safety assessment, the importance of confidence building in the applied models, codes and input data is paramount, especially since the involved timescales are extremely long. To that aim, quality procedures are introduced which demand the application of model validation (whenever possible), model qualification and code verification. The latter is typically implemented by doing benchmarking analyses in which numerical codes are compared with others or, in case of simplified systems, with analytical solutions. Recently, COMSOL Multiphysics has been successfully applied in an internationally organized benchmarking exercise in the framework of the European project PAMINA. This paper discusses some highlights of the results obtained by SCK•CEN with the Finite Element (FE) tool COMSOL Multiphysics v.3.2 (COMSOL, 2005) and corresponding results with the Finite Volume (FV) code PORFLOW v3.07 (Runchal, 1997).

2. Benchmark Definition

2.1 General Approach

The benchmark focused on a hypothetical repository in a very low-permeable clay layer for which the evolution of radionuclide activity fluxes to an overlying aquifer were to be calculated, as well as the evolution of the radionuclide concentration in the source zone. The physical processes included in the model are radioactive decay and production of daughter nuclides, the combination of a flux-type source in the waste zone with a solubility limit and the associated build-up of precipitate, advective-diffusive transport through the different porous media and linear reversible sorption on the clay particles.

2.2 Description of the Repository System

The repository system considered in this benchmark is based on the French repository design for disposal of vitrified high-level waste (HLW) in clay, as described in "Dossier 2005 Argile" (ANDRA, 2005).

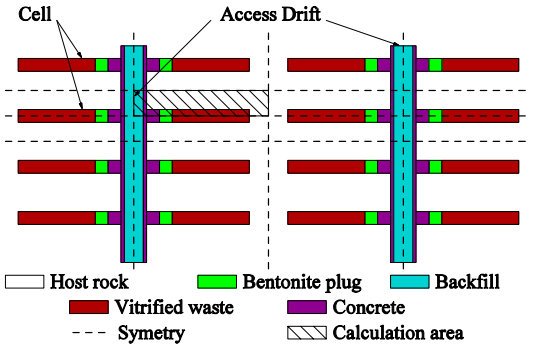

Figure 1. Top view of disposal area part including symmetries and reduced calculation area.

Figure 1 shows a schematic overview of the disposal area located at the midplane of a 100 m thick clay layer, consisting of a series of cylindrical disposal drifts, connected to an access drift. The vitrified waste is inserted in the disposal drift, which is sealed with a bentonite plug and a concrete plug. After emplacement of the waste, the access drift is filled with some backfill material. The 3D rectangular approximation of one disposal cell, used as basis for the calculations described in this paper, is depicted in Figure 2.

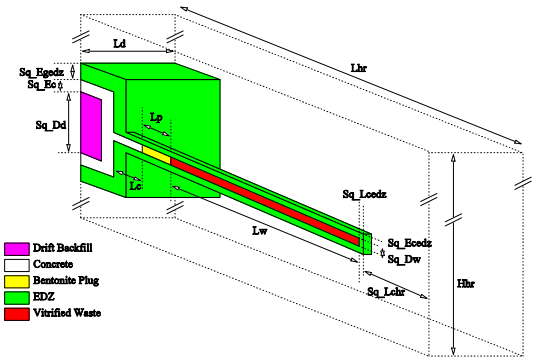

Figure 2. Rectangular geometry approximation of a disposal cell for vitrified HLW in clay. "EDZ" stands for Excavation Disturbed Zone.

2.3 Conceptual Model and Model Geometry

The governing equation for transport of radionuclide i in saturated media is stated as:

$$(\theta + \rho_b K)\frac{\partial c_i}{\partial t} + \nabla\left[-\theta \mathbf{D_i}\nabla c_i + \mathbf{u}c_i\right] = R_{Li} + R_{Pi} + S_{ci}$$

where c_i is the solute concentration in the liquid phase, K is the distribution coefficient between the solid and the liquid phase, θ is the porosity, ρ_b is the bulk density, $\mathbf{D_i}$ is the hydrodynamic dispersion tensor and $\mathbf{u}$ is the vector of flow velocities. R_{Li} and R_{Pi} represent reactions (such as radioactive decay and production, or the precipitation/dissolution process) in the liquid and solid phase respectively and S_{ci} denotes a solute source. Since the hydraulic gradient (1m/m in upward direction) over the clay formation is quite high, advection cannot be neglected, even though the hydraulic conductivity of the clay is very small (K_z=10^{-13} m/s). As such, radionuclide transport is both governed by diffusion and advection.

The release of radionuclides into the pore water of the repository system is determined by the dissolution of the vitrified waste. Water will first reach the vitrified waste when the containment of the steel canister fails, which is assumed to occur 4000 years after waste emplacement. Thereafter dissolution of the glass and the release of the embedded radionuclides is assumed to go on for 100 000 years until the whole matrix is dissolved. The release of some radionuclides, however, will be controlled by their solubility.

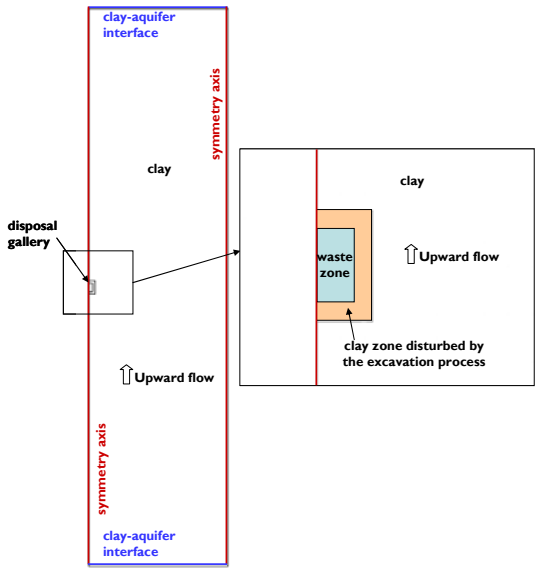

Figure 3. Schematic description of the radionuclide transport problem.

The model geometry of the radionuclide transport problem is schematized in Figure 3. Note that, through the use of symmetry planes, only half of 1 disposal gallery needs to be modelled. At these symmetry planes, zero-flux boundary conditions are applied. At the clay-aquifer interface, an infinite dilution (C=0) boundary condition is applied. The source is located in the waste zone, surrounded by a so-called excavation disturbed zone (EDZ), with slightly different properties.

In this paper, benchmark results will be presented for ^{129}I, ^{79}Se and the radionuclide chain 4N+1. Detailed listings of input data (radionuclide transport properties in the different materials, radionuclide inventory data, and solubility limits of the considered radionuclides) can be found in (CEA, 2008). We assumed that the grid spacing for both the FE and FV codes was sufficiently fine to obtain an accurate solution and there has been no attempt to use coarser grids to assess accuracy degradation.

3. Results and Discussion

As output parameters, we systematically compared the radionuclide flux to the overlying aquifer, which in safety assessment usually serves as basis to derive potential doses via a specific biosphere pathway, and the concentration in the source zone.

3.1 Results for ^{129}I

^{129}I is transported through the clay layer by advection and diffusion. It is highly soluble and because it migrates as an anion, it does not adsorb on the negatively charged clay particles. ^{129}I is thus fairly mobile in the clay. Both COMSOL (FE) and PORFLOW (FV) calculations were performed with stepwise increasing complexity in the transport processes. Results for the cases where diffusion is the only solute transport process are shown in Figure 4 in terms of activity fluxes to the overlying aquifer per disposal cell. Figure 4 shows an excellent agreement between the 2D PORFLOW and COMSOL results with a slow dissolution of the vitrified matrix (red curves).

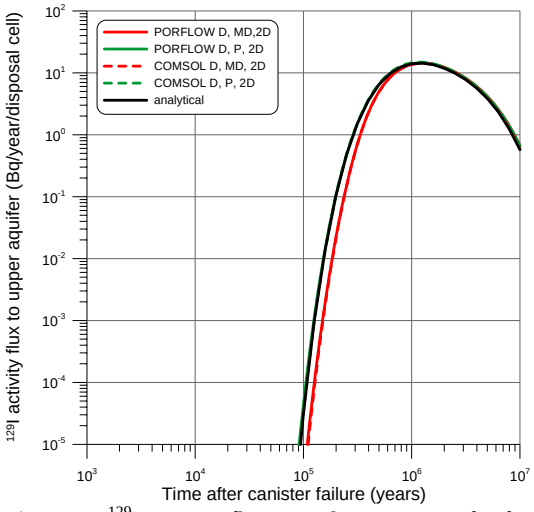

Figure 4. ^{129}I activity fluxes to the upper aquifer for the 2D diffusive (D) transport calculations. (MD: release determined by matrix dissolution)

If we consider an instantaneous release of the total ^{129}I inventory, this problem can be easily solved analytically, which is shown by the black curve. For completeness, 2D PORFLOW and COMSOL results using a pulse-type source term are shown as well. They match perfectly to the analytical solution. It can be seen that the clay host formation buffers the instant release of ^{129}I so effectively that the differences between a pulse-type release and slow matrix dissolution are marginal.

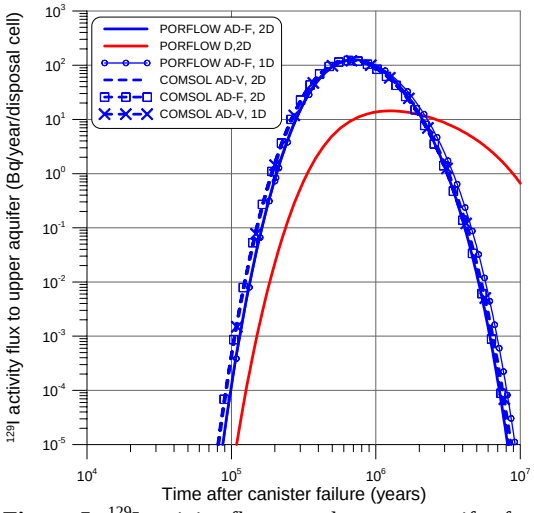

Figure 5. ^{129}I activity fluxes to the upper aquifer for 1D and 2D advective-diffusive transport calculations. (AD-F: flow equations are solved; AD-V: flow implemented through a fixed flow field)

Upward flow results in higher activity fluxes and a shorter transport time into the overlying aquifer compared to diffusion alone (see Figure 5). All COMSOL calculations show the same results: there is no remarkable difference in dimensionality (1D vs 2D) or solving the flow equations or imposing a fixed flow field. The increase in ^{129}I flux occurs slightly sooner in the COMSOL calculations compared to PORFLOW, but the peak flux is the same for all cases. PORFLOW results in 1D become less accurate after 10^6 years (probably because of different numerical dispersion between the models).

Figure 6 shows the ^{129}I concentration in the source region for the different cases, with excellent agreement between both codes. At the source, there is a gradual accumulation of ^{129}I until the vitrified matrix is completely dissolved (at 10^5 years). Thereafter, the concentration decreases because of solute transport away from the source.

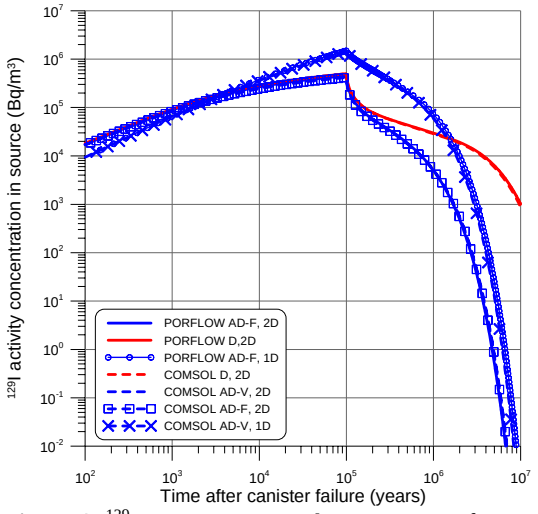

Figure 6. ^{129}I concentration in the source zone for 1D and 2D advective-diffusive transport calculations.

For the advective flow cases, the concentration decreases faster after 10^5 years, because the source region is "washed out". This effect is already visible at the end of matrix dissolution, where concentrations reached at the source zone are slightly lower. Off course, the 1D calculations do not accurately represent the source concentration. In the beginning the source concentration is too low because the solute is diluted in a unit thickness of the second dimension, and later on the concentration is too high because the radionuclide can only be transported in one dimension (underestimation of the "loss" by migration in the second dimension). At the level of the aquifer however, the concentrations and fluxes match the 2D results very well, as could be seen in Figure 5.

3.2 Results for ^{79}Se

^{79}Se migrates as an unsorbed species. When the vitrified waste matrix slowly dissolves, the concentration of Se will build up until its solubility is reached. From that moment, excess Se that dissolves from the waste will precipitate resulting in a constant ^{79}Se concentration in the waste zone. In the FV model PORFLOW 3.07, it is not readily possible to start from a flux-type source term (as is the dissolution of the matrix) and switch to a constant concentration-type source term when the solute's solubility is attained. As such, the approach closest to reality in PORFLOW is to have an instant release of the solute's inventory up to the concentration limited by solubility, with the excess inventory present as precipitate. In general, this approach is not so far from reality since the inventory and initial release rate are high enough for the solubility limit to be rapidly attained.

In COMSOL, the combination of slow matrix dissolution and solubility controlled phases is possible by implementing the precipitate as a separate species and defining proper exchange reactions, representing the precipitation and dissolution processes, between the two species. This approach is indicated by "MD-SL" in the figures (matrix dissolution followed by solubility limited release).

Figure 7 shows the excellent match between the COMSOL and PORFLOW calculations both for the diffusive (red) cases and the advective-diffusive (blue and green) cases. In Figure 8, the COMSOL MD-SL calculation shows that the solubility concentration of Se is quickly reached (after about 2 years) and this more sophisticated release model has no measurable effect on the resulting ^{79}Se fluxes to the aquifer, as shown in Figure 7. Figure 9 shows the precipitate "concentration" (amount present per unit volume) for the COMSOL SL and MD-SL case. Obviously, the precipitate builds up more slowly in the MD-SL case, since the majority of the ^{79}Se is still embedded in the matrix.

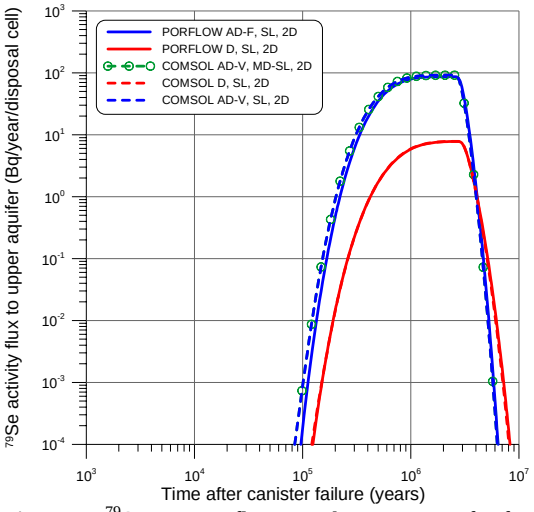

Figure 7. ^{79}Se activity fluxes to the upper aquifer for the 2D advective-diffusive transport calculations.

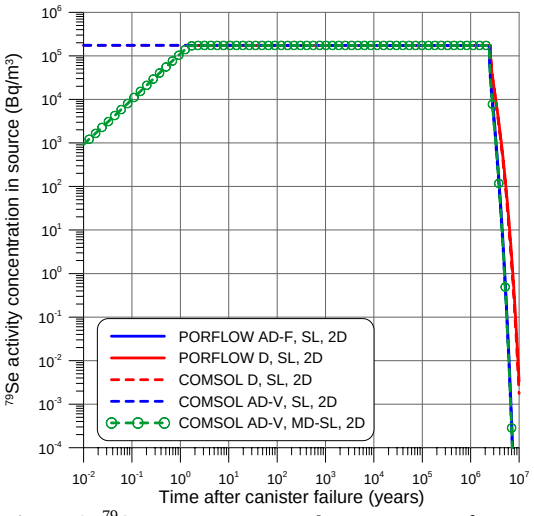

Figure 8. ^{79}Se concentration in the source zone for 2D advective-diffusive transport calculations.

3.3 Results for the 4N+1 radioactive decay chain

In this benchmark, the 4N+1 actinide chain will be represented by the following 3 members with relatively long half-life: ^{237}Np → ^{233}U → ^{229}Th. The transport of these radionuclides is characterised by very strong sorption on the clay particles. However, since the emphasis in this benchmark was on evaluating the feasibility of the implementation of important processes, only mild sorption was applied for these radionuclides, not representing as such realistic conditions.

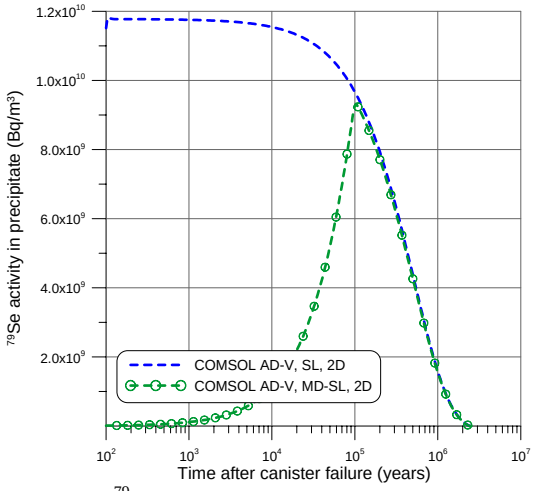

Figure 9. ^{79}Se amount present as precipitate in the source zone for the COMSOL SL and MD-SL case.

It may be noted that in COMSOL, there is considerable flexibility in modelling a reaction chain. The number of chain members is unlimited but the exchange (decay) reactions are to be written by the user. In PORFLOW 3.07, the number of chain members is limited to 4 (which is usually sufficient for the conventional radioactive waste types). Furthermore, the release of ^{237}Np is limited by its solubility. As explained before, the most realistic release model is the MD-SL case (which is only possible in COMSOL). Likewise, the concentration of the daughter product ^{233}U, continuously generated by radioactive decay, could exceed its corresponding solubility, at which point the build-up of precipitate should occur. This effect was taken into account in one of the COMSOL calculations "MD-SL(Np,U)". Another issue concerns the inventory of the different actinides. "I(Np)" means that only the initial activity of ^{237}Np is implemented as the source term, and its daughters are generated by radioactive decay, while the "I(Np,U,Th)" calculations take also into account the activity of ^{233}U and ^{229}Th initially present in the waste. The complexity (realism) of the source term description was gradually increased, taking into account these phenomena step by step.

The ^{237}Np fluxes for the considered chain members are shown in Figures 10 to 12. All COMSOL (FE) calculations give a slightly earlier arrival of the solute plume at the aquifer level compared to PORFLOW (FV), but differences are small. Additional realism in the

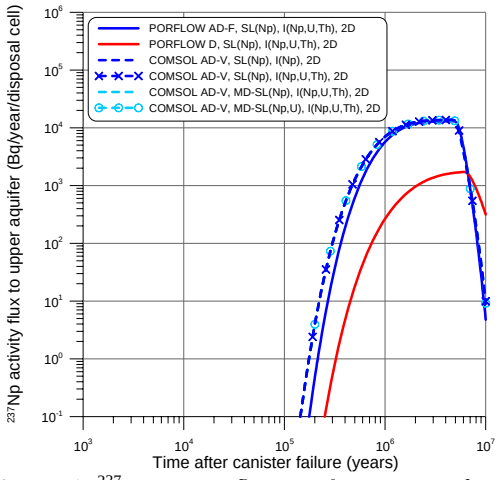

Figure 10. ^{237}Np activity fluxes to the upper aquifer.

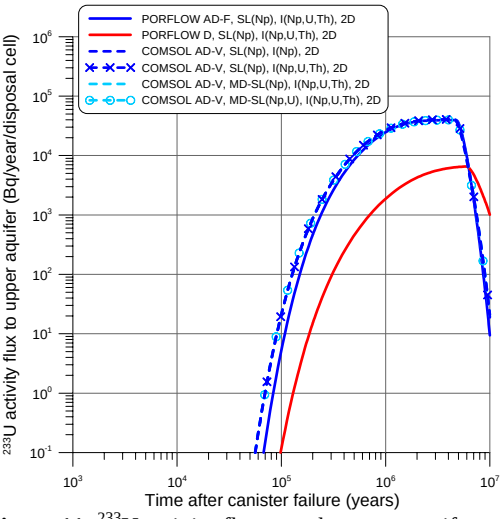

Figure 11. ^{233}U activity fluxes to the upper aquifer.

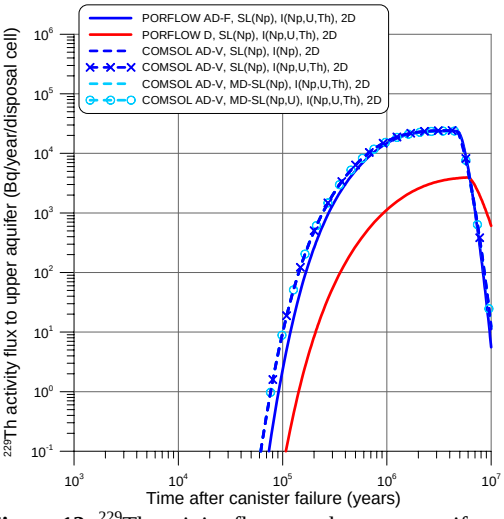

Figure 12. ^{229}Th activity fluxes to the upper aquifer.

source term formulation (initial inventories of daughter products, realistic dissolution/ precipitation of Np and/or U) does not influence the fluxes to the overlying aquifer. However, these effects are clearly visible when plotting the source concentrations (see Figure 13 to 15). The "MD-SL" cases show that the solubility of ^{237}Np is attained about 10 years after the start of matrix dissolution (Figure 13), while the "MD-SL(Np,U)" case shows that the U solubility is only reached after some hundreds of thousands of years (Figure 14). This can be more clearly seen in Figure 16, where the ^{233}U activity in the precipitate is plotted.

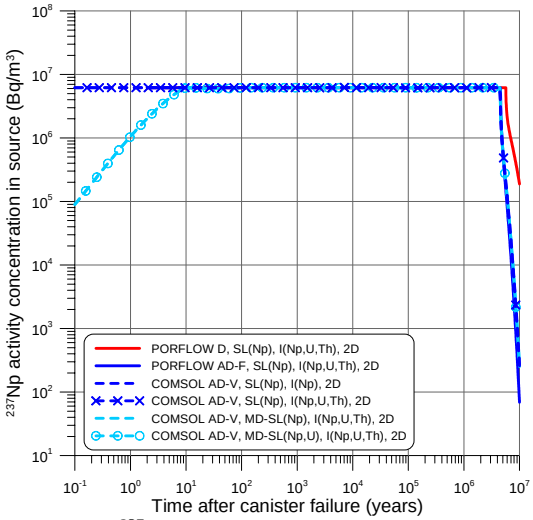

Figure 13. ^{237}Np activity concentration in the source zone.

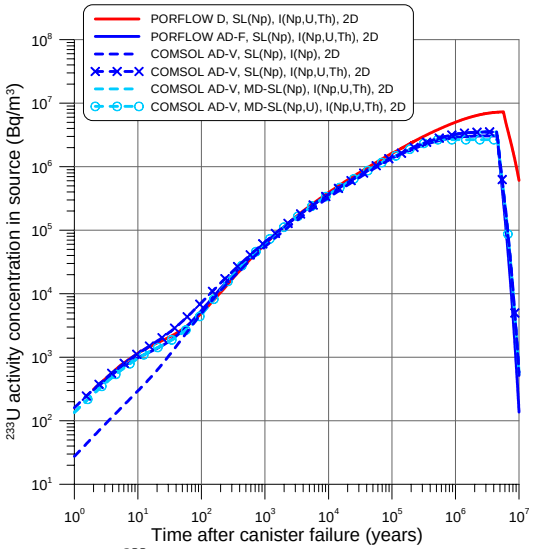

Figure 14. ^{233}U activity concentration in the source zone.

Furthermore, it is clear from Figures 14 and 15 that the initial activity of the daughter nuclides is negligible compared to the activity that is produced by radioactive decay.

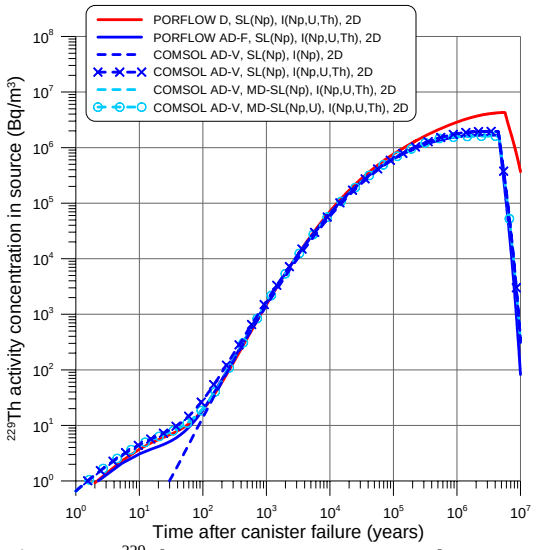

Figure 15. ^{229}Th activity concentration in the source zone.

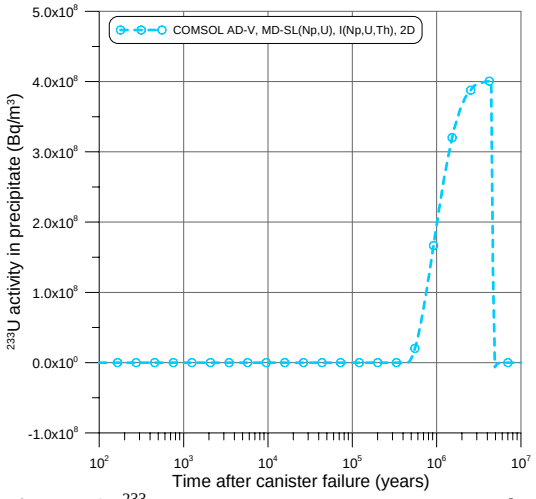

Figure 16. ^{233}U amount present as precipitate in the source zone.

It is worth mentioning that some caution is required while implementing a solubility-limited source term. At the moment of precipitate depletion, which can occur after some hundreds of thousands to millions of years, timesteps are usually already very large (in most cases, large timesteps are possible without compromising the stability as the system is close to steady state, or at least not experiencing sharp gradients). Because of the occurrence of such a discrete event at late times, it might be possible that too much precipitate is put into solution in one timestep, possibly resulting in negative precipitate concentrations (noticeable in Figure 16) and mass-balance inaccuracies.

7. Conclusions

COMSOL Multiphysics can be a valuable complementary modelling tool for safety calculations of a radioactive waste repository. COMSOL has shown excellent flexibility in adding more realism in the mechanisms of radionuclide release. Although this more realistic description of the source term has no effect on the resulting fluxes to the aquifer, these type of calculations are nevertheless of important to gain insight in how the repository system works (so called performance assessment calculations) and why it is safe. The very good agreement between the two codes (considering the same conceptual model) indicates that the implemented governing equations are accurately solved, *i.e.* code verification. This is of importance in building confidence in the results of a safety assessment, especially considering the extremely slow processes, and hence long timescales involved.

8. References

1. Runchal, A.K, PORFLOW, a Software Tool for Multiphase Fluid Flow, Heat and Mass Transport in Fractured Porous Media. User's manual, Version 3.07. ACRi, Bel Air, US (1997)
2. COMSOL, COMSOL Multiphysics 3.2, Earth Science Module Model Library, COMSOL AB, Stockholm, Sweden (2005)
3. ANDRA, Dossier 2005 Argile, Châtenny-Malabry, France (2005)
4. CEA, PAMINA deliverable of WP4.2, PA approaches based on different geometric complexity of modelling, EC, to be published

9. Acknowledgements

This benchmark exercise was undertaken in the framework of the PAMINA project ("Performance Assessment Methodologies in Application to Guide the Development of the Safety Case"), a project under the auspices of, and with the financial support of the EC (6th FP).