Building a Robust Numerical Model for Mass Transport Through Complex Porous Media

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Abstract: When modelling complex physical problems model simplifications present an important step in a modelling procedure. One of the most common simplifications is dimensionality reduction. With dimensionality reduction the model becomes better manageable and consequently more robust. Mass transport modelling through porous media is typically characterized by complex physics and geometry. In the particular case of radionuclide transport modelling for radioactive waste repositories, an additional level of complexity, and thus uncertainty, originates from the long time frames involved. Performing a safety analysis of a radioactive waste disposal system requires therefore reasonable simplifications which enable us to model such complex system thereby minimizing the effect of uncertainties. In this work several examples, calculated by COMSOL Multiphysics 3.2, are selected to show the acceptable level of simplification to assure representative and yet robust modelling of radionuclide transport from a disposal facility.

Keywords: Model simplification, model dimension, radionuclide transport, porous media flow and transport, mass release mechanisms.

1. Introduction

The long-term safety of radioactive waste disposal facilities is usually demonstrated with the support of a safety assessment. This normally includes modelling of radionuclide release from a multi-barrier near-surface or deep repository to the geosphere and biosphere [1]. The overall system is characterised by an enormous complexity of interconnected physical-chemical phenomena acting on a wide range of length scales. In addition to physical-chemical and geometrical complexity, the simulation of long time frames in the order of tens of thousands of years are required to calculate release of strongly sorbed radionuclides from a concrete based disposal facility to the environment (i.e. groundwater and biosphere). During this long period most radionuclides decay within the facility. The impact of the release of a residual fraction of radionuclides on human health is evaluated by means of numerical modelling. The numerical problem is usually simplified by dividing the disposal system into three linearly dependent components: i) near-field where leaching of radionuclides from the engineered disposal facility to the groundwater takes place, ii) geosphere and iii) biosphere where subsequent use of groundwater for drinking, irrigation of fields and watering cattle is accounted for [2]. In this paper we focus on the near-field safety assessment of a near-surface disposal facility. The near-field comprises concrete waste disposal containers, termed monoliths, and structural components of the vault, termed modules, such as concrete walls, floor and roof, and a multi-layer cover isolating the disposal facility from the environment. The monoliths are composed of the conditioned waste, cementitious backfill and a concrete box, the container. Usually a large variability exists in the geometry and size of conditioned waste packages: most common are cylindrical drums, but also bulk waste will be present. Models used for the long-term safety assessment should accommodate these variations in different packages, but on the other hand should remain numerically efficient and practical, in terms of input parameters. Therefore the real system is simplified considerably, providing that each simplification shall not underestimate the impact, i.e. should be conservative. One of the most commonly used simplifications is the reduction of dimensionality as shown in Figure 1.

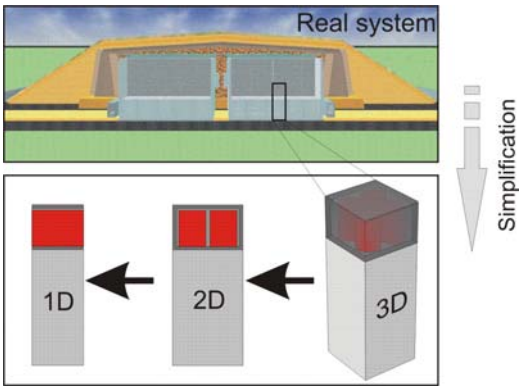

Figure 1. Simplification of the 3D domain into a 3D, 2D and 1D model.

In order to demonstrate whether the simplification to a very simple 1D model is adequate, several steps are performed. The adequacy is tested for several different packaging types and for different mass release mechanisms, namely instantaneous, diffusional and dissolutional release [3]. The evaluation is based on mass fluxes (here radionuclide fluxes) across the bottom of the model. The full multi-monolith model is simplified to include only the bottom monolith and filled inspection rooms in order to limit the large number of elements in 3D.

2. Physical background and modelling approach

2.1 Source geometry

The problem presented involves solute transport of decaying and sorbing substances (i.e. radionuclides) in saturated porous media. One of the challenges in radionuclide transport modelling is how to properly deal with the large difference between often very high initial concentrations in the source zone and output concentrations, which may be over 10 orders of magnitude different. In such case mass transport calculations by diffusion and/or advection often suffer from negative concentrations or oscillation which lead to physically unrealistic results. Thus modelling of radionuclide transport requires a carefully defined model, especially when high Peclet numbers (advection dominates over diffusion) are involved. One possible solution to oscillations is the use of logarithmic concentrations, but this turns out not to be a good solution because when the oscillation occurs, it drives concentrations towards 0 or logarithmic concentrations towards -∞. At some point in the simulation the concentration value becomes too low to evaluate the exponential term. In addition, automatic time stepping determination can fail in some cases. Another solution is applying a denser grid in regions experiencing the highest gradient in combination with a smoothed Heaviside function *flc2hs* [4] instead of a step function for the high initial concentrations in the source zone. Yet another possible solution, not tested here, is the application of streamline diffusion [4].

Due to a much lower geometrical complexity of 1D and 2D models over 3D models, both grid refinement and smoothing of the source zone are much easier to achieve. For this reason it is beneficial to have the model dimension as low as possible. In this paper we tested the adequacy of 2D and 1D models derived from more realistic 3D models under advection dominated flow conditions for three different source release mechanisms. The 3D geometry for evaluating the two source geometries (i.e. five 220-*l* drums or 400-*l* drums) is presented in Figure 2.

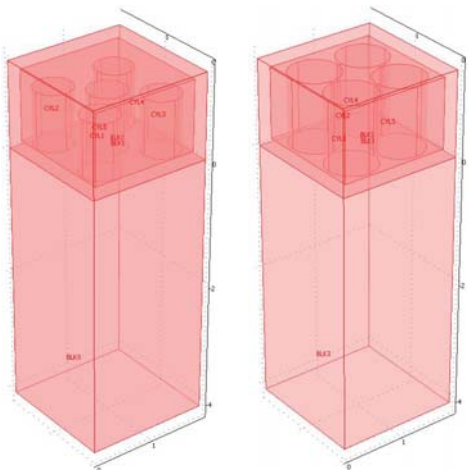

Figure 2. 3D model geometry for 220 *l* drums (left), 400 *l* drums (right). Total height of the domain is 5.34 m, horizontal dimensions are 1.94x1.94 m².

In case of the 3D geometry, each drum is put at an initial condition (i.e. initial concentration)

of $C(t_0)$. The initial source concentration in the liquid phase is defined as:

$$C(t_0) = \frac{A}{V \cdot R \cdot \eta}, \quad (1)$$

where A is the initial activity, arbitrary set to 10^4 Becquerel (Bq), V is the volume of the source region, R is the retardation coefficient and η is the porosity of the source material – here assumed to be a cementitious matrix. Smoothing of the initial source concentration (here for 220 *l* and 400 *l* drums) is performed in all dimensions. The smoothing half-interval span is 3 cm. An example of the smoothing function in equation (2) is given for the 3D geometry with 400 *l* drums, where z_{bot} and z_{top} represent bottom and top drum coordinates, respectively, and x_n and y_n are drum ground view center coordinates.

$$\begin{aligned}
smooth_z &= flc2hs(z - z_{bot}, 0.03) + (1 - flc2hs(z - z_{top}, 0.03)) - 1 \\
smooth_1 &= 1 - flc2hs(\sqrt{(x - x_1)^2 + (y - y_1)^2}) - r, 0.03) \\
smooth_2 &= 1 - flc2hs(\sqrt{(x - x_2)^2 + (y - y_2)^2}) - r, 0.03) \\
smooth_3 &= 1 - flc2hs(\sqrt{(x - x_3)^2 + (y - y_3)^2}) - r, 0.03) \\
smooth_4 &= 1 - flc2hs(\sqrt{(x - x_4)^2 + (y - y_4)^2}) - r, 0.03) \\
smooth &= smooth_z \cdot (smooth_1 + smooth_2 + smooth_3 + smooth_4)
\end{aligned} \quad (2)$$

As a result of the smoothing operation, the volume occupied by the source zone becomes smaller. Therefore, the volume (which is used to calculate the concentrations from the initial activity in equation (2)) is calculated as given in equation (3).

$$V = \int_V smooth \, \mathrm{d}V . \quad (3)$$

To counterbalance the volume change and to assure a correct mass balance, the initial concentrations are multiplied by the smoothing function as well as: $C(x,y,z)=C_0 \cdot smooth(x,y,z)$, where C_0 is the concentration calculated on the basis of the initial activity and calculated volume V through equation (3).

Concentration boundary conditions are the same in all calculation cases. At the top a zero concentration (C=0) boundary condition is imposed, the bottom boundary condition is an advective flow boundary condition (equivalent to zero gradient), and all other boundaries are no-flow (Neumann) boundary conditions. Different modelling approaches are required for different release mechanisms as will be discussed in the next sections.

2.2 Instantaneous release model

Instantaneous release assumes all source activity is released instantaneously and completely. This is the most conservative release model one can assume for modelling radionuclide release from a waste disposal facility. Radionuclides are leached from the conditioned waste by infiltrating water and by diffusion. To facilitate water flow and mass transport simulations through concrete components, a very conservative approach is adopted in this paper imposing a maximum and uniform vertical water flux across the facility. This would correspond to physically degraded concrete components, which will be the ultimate state of the concrete in the long run. This assumption turns the problem into a simplified advection-dispersion driven system, with equilibrium mass exchange between solids and pore-water. In other words, a transient period with gradually degrading concrete and consequently a gradually increasing water flux is neglected. The mass exchange or sorption process is the consequence of the assumption that concrete will keep high sorption properties for a very long time. The same assumption of uniform water flow is used for all dimensions. Two different waste package geometries in a monolith are used for comparison here, i.e. 220 *l* and 400 *l* drums. Two radionuclides, ^{36}Cl and ^{129}I with different half-lives $T_{1/2}$ and sorption behaviour (K_d) are used in the comparison (see Table 1).

Table 1: Half-lives and partitioning coefficients for ^{36}Cl and ^{129}I.

	^{36}Cl	^{129}I
K_d [l/kg]	100	1
T_{12} [y]	301000	16100000

2.3 Diffusional release model

Diffusional release takes place when the release of radionuclides is limited due to diffusion through a porous waste form such as cement (e.g. cement-conditioned waste form) or bituminized waste. Water flow through the waste form is considered negligible because the waste form is intact and therefore has a low

permeability. Modelling approaches for each dimension are presented in Figure 3.

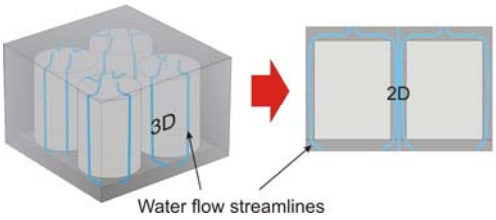

Figure 3. Diffusional release models for 3D and 2D.

Compared to Figure 2, the model includes only one monolith without concrete base for the reason of computational efficiency. The momentum and mass conservation equations are still solved decoupled. Initially, only Darcy's porous media flow is calculated in steady-state using a flux boundary condition at the top and a pressure boundary condition at the bottom. Hydraulic conductivity of the waste region (k=10^{-12} m/s) is low enough to satisfy diffusion driven transport. After obtaining steady-state water flow the transport equation is calculated. The application of a diffusion-limited source zone surrounded by advection-driven transport in 1D is not straight-forward and therefore 1D calculations have not been performed.

2.4 Dissolutional release model

A dissolutional release mechanism is applied when mass release is controlled by the time-dependent dissolution of the waste form. From a geometrical perspective, the dissolutional release propagates from the waste form surface inward. Examples of dissolutional release are corrosion of metal parts or dissolution of polymers or vitrified waste forms [5]. From a modelling point of view, the parameter that defines radionuclide fluxes from the source towards the surrounding concrete components is the dissolution rate δ. In the case described below, dissolution is applied to 400 l drums containing polymerized waste. Due to dissolution, radius $r(t)$ and height $z(t)$ of the waste zone and consequently also reactive surface area $A(t)$ decrease with time as described in equation (4).

$$r(t)=\begin{cases} r_0-\delta t & r_0 \geq \delta \cdot t \\ 0 & r_0 < \delta \cdot t \end{cases}$$
$$z(t)=\begin{cases} z_0-\delta t & z_0 \geq \delta \cdot t \\ 0 & z_0 < \delta \cdot t \end{cases} \qquad (4)$$
$$A(t)=2\cdot\pi\cdot r(t)\left(r(t)+z(t)\right).$$

Modelling approaches in 3D/2D and 1D are very different. In 3D/2D the dissolution model is characterized by a shrinking source term. For example, in case of a drum, the drum diameter and height decrease with time. Along with the change of source dimensions, the water pattern changes as well, as the initially low permeability materials will be replace by more porous and more permeable ones. This is incorporated in a fully coupled momentum and mass conservation equation. Initially steady-state Darcy's water flow is calculated. The solution is then used as an initial conditions for time dependent calculations. Because hydraulic conductivity of the waste form is arbitrarily taken to be 4 orders of magnitude lower than the hydraulic conductivity of the concrete backfill and container (10^{-8} m/s), logarithmic averaging has been made between the regions with contrasting conductivities:

$$e^{\ln k_0 (1-smootht)+\ln k_1 \cdot smootht}, \qquad (5)$$

where k_0 and k_1 are hydraulic conductivities of concrete and waste form, respectively. Smoothing function *smootht* is similar to equation (2), but with the time dependent parameters $z(t)$ and $r(t)$ instead of z and r.

In 1D, the dissolution is described by a much more simple model, that directly calculates the flux from the waste region. The flux, F_C, is calculated as

$$F_C[Bq/y]=C_0\cdot\delta\cdot A(t)\cdot e^{-\ln(2)/T_{1/2}\cdot t}, \qquad (6)$$

where $T_{1/2}$ is half-life of radionuclide, all other parameters are as defined previously. Water flow is uniform over the whole domain. To reduce the computational time, a relatively high dissolution rate δ of 100 µm/y is assumed, by way of example, resulting in a fast leaching.

Water flow boundary conditions are a defined flux at the top boundary and constant pressure at the bottom boundary. All other boundaries are no-flow boundaries. In 3D the solver settings had to be changed from default

ones in order to obtain convergence. Conjugate gradients with algebraic multigrid preconditioner were used to iteratively solve the system of equations. The preconditioner quality parameter was decreased to 2 and relative and absolute tolerances were 0.0001 and 0.00001, respectively.

3. Numerical results

The numerical results are discussed according to the three release mechanisms used. Model sensitivity to geometrical issues is mainly demonstrated using instantaneous release, while diffusional and dissolutional release mechanism results are used to demonstrate the limits of model simplification. Output from the near-field calculations is through radionuclide fluxes across the bottom of the calculation domain, where the radionuclides enter groundwater.

3.1 Instantaneous release

Because the instantaneous release model is very simplified, assuming unidirectional water flow everywhere, the differences between output fluxes from all models for a specific waste package prove to be negligible as presented in Figure 4. The impact of using different waste packages (i.e. 200 or 400 *l* drums) is small too (Figure 4): the 200 *l* drums produce slightly higher fluxes than the 400 *l* drum.

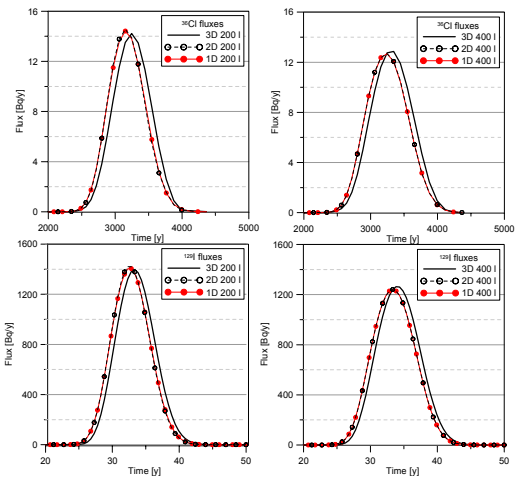

Figure 4. Flux comparison in 3D, 2D and 1D for ^{36}Cl and ^{129}I for instantaneous release.

Correlation analysis between the maximum fluxes and height of waste source shows, based on 6 different packaging types (only 220 *l* and 400 *l* are shown in this paper), that the differences in peak flux between different source geometries are perfectly correlated to the height of the package and not to their volume. This behaviour is based on the orientation of the water flow in z-direction, causing the transport to be dominantly oriented downwards. For this reason the results in lower dimensions are in very good agreement with the 3D results.

3.2 Diffusional release

From Figure 5 one can observe the relatively good agreement between the 3D and 2D model in case of diffusional release. Their peak fluxes are nearly identical, but the 2D peak occurs earlier than the 3D peak because of initial concentration smoothing and consequently lower concentrations on the waste/concrete interface for the 3D versus 2D model. The results in Figure 5 are presented for ^{36}Cl and the 400 *l* drum.

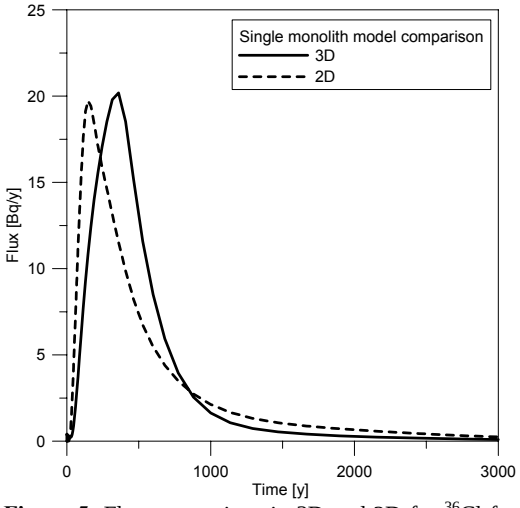

Figure 5. Flux comparison in 3D and 2D for ^{36}Cl for diffusional release.

Note that the level of dimensionality reduction for diffusional release must be checked for each case separately because the diffused mass depends on the diffusive surface available.

This condition sometimes cannot be appropriately accounted for in 2D.

3.3 Dissolutional release

Dissolutional release presents the most complicated case from the modelling point of view. This is not only due to the complicated procedure for creation of a fully coupled 3D shrinking model, but also due to the different ways the source term is implemented in different dimensions, e.g. initial condition concentration source in 3D/2D and flux source in 1D. Graphical presentation of a shrinking source term in 3D at three time steps is given in Figure 6.

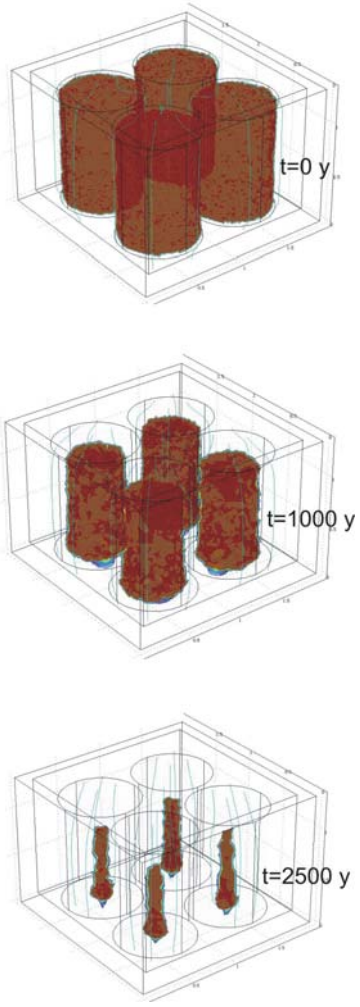

Figure 6. Shrinking source term and streamlines for dissolutional model at three time steps.

As a result of the shrinking source term, the flow pattern in the monolith changes as the permeability of waste and conditioning matrix increase. A comparison of results between 1D and 3D is seen in Figure 7.

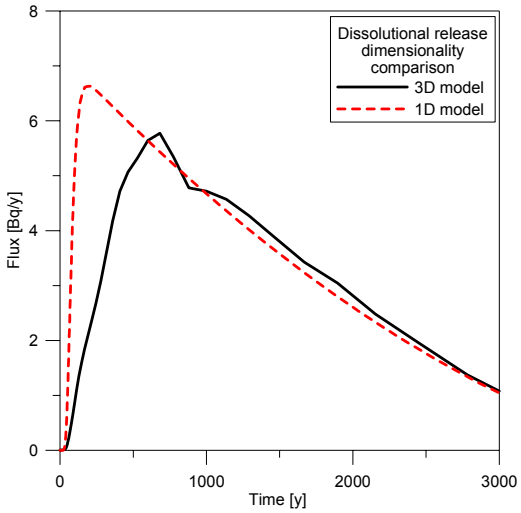

Figure 7. Flux comparison between 3D and 1D model for dissolutional release (400 *l* drums).

The difference in peak flux for ^{36}Cl is approximately 15%, where 1D is larger than 3D. This difference is probably due to the smoothing of concentrations in 3D, which makes the initial leached concentrations somewhat lower (at the edge of the waste region at the initial time). Based on the observation that the 1D model is conservative, the use of a 1D model is fully justified. Also note that the 1D model is much more efficient than the 3D, i.e. calculation times[1] for the 1D model is 5 seconds whereas it is 19 hours for the 3D model.

In contrast with the diffusional release model, for which the simplification from 3D to 1D cannot be made, the simplification is justified for dissolutional release, because geometrical properties of the 3D release model are taken into account through the use of release fluxes that are input into the 1D model.

[1] INTEL CORE2 CPU 6700 @ 2.66 GHz

4. Conclusions

Results presented in this work show that the use simplified and thus more robust models can be justified for modelling of radionuclide release from conditioned waste packages granted in concrete containers. The models in lower dimensions can accommodate much less numerical uncertainties as well as physical uncertainties on, for example, migration processes, type of waste forms or water flow provided that the results obtained with lower dimensional models are conservative. However, the automatic use of a 1D model is in some cases not justified as shown for example in case of diffusional release. Therefore it is important to test each different type of simplified model against more realistic 3D models before using them in real safety analysis. In the latter case a 2D model is the minimum required model dimensions.

5. References

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