Preliminary investigation of radionuclide release under two-phase conditions from a proposed L/ILW repository in Switzerland

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Introduction

The Swiss National Cooperative for the Disposal of Radioactive Waste (NAGRA) is investigating the feasibility of siting a low- and intermediate-level radioactive waste (L/ILW) repository in subsurface geological environments. The design of the L/ILW repository consists of a horizontally accessed cavern system located in a low-permeable host rock. One of the issues in the safety analysis is the effect of gas generation (primarily hydrogen) from anaerobic corrosion and degradation of waste material. The development of a free gas phase can result in the displacement of contaminated pore waters out of the repository.

In a previous study (Senger et al. 1994) the effect of gas generation on pressure buildup in backfilled caverns was investigated. It was concluded that, inititally, gas dissolves until gas saturation in the pore water is reached. Thereafter, a free gas phase is established coinciding with a pressure increase and a concomitant porewater displacement out of the caverns into the geosphere. These processes are mainly controlled by the following parameters: gas generation rate, intrinsic permeability of the host rock, relative permeability, and initial gas saturation.

For the investigation of the effects of gas-related phenomena on radionuclide transport preliminary near field calculations were carried out using TOUGH2/EOS7R (Oldenburg & Pruess 1996a,b) with the objectives of i) comparing the calculated radionuclide release rates under fully saturated conditions (no gas generation) with results from NAGRA's standard L/ILW near field code SEFTRAN (NAGRA 1994), and ii) investigating the release of non-volatile radionuclides under two-phase conditions, taking gas generation into account.

Modeling approach

The model domain is represented by a 2-D vertical cross section through a single L/ILW repository cavern and the adjacent host rock (Figure 1). The 2-D model is oriented perpendicular to the cavern axis, incorporating the engineered barriers of the repository (waste packages, container backfill, container walls and lid, cavern backfill, liner) and the host rock (disturbed zone around the cavern, undisturbed host rock). The model domain has an extension of 140 m horizontally and 160 m vertically.

The finite element mesh used in the simulations of radionuclide release under saturated conditions with NAGRA's standard near field transport code SEFTRAN is shown in Figure 2. The rectangular mesh consists of 4'320 elements grouped into 8 hydraulic units. As can be seen in Figure 2, some of the engineered barriers are very thin (e.g. thickness of container walls is 0.2 m) and are represented by only one row of elements. To test potential grid effects, a finer finite element grid with 14'672 elements was used. In the case of radionuclide transport simulations with TOUGH2/EOS7R only the centres of the elements of the original coarse grid are taken into consideration for the finite difference grid.

When neglecting the gas generation in the caverns, the release of radionuclides from the near field into the geosphere is driven by the external hydraulic gradient associated with the regional groundwater flow system at the proposed site for the L/ILW repository. When gas generation is taken into account, a free gas phase is established. In this case the radionuclide release out of the caverns is driven by two processes: i) the pressure build-up due to gas generation, which leads to the displacement of contaminated pore water out of the cavern, and ii) the external hydraulic gradient.

A selected set of parameters used for the simulation of radionuclide release is summarised in Table 1. Both the engineered and natural barriers are characterised by material dependent properties such as absolute permeability, relative permeability, porosity, pore diffusion constants, mass of sorbing material, and distribution coefficients. The absolute permeabilities within the engineered barriers range from 10^{-15} m^2 for the backfill material (coarse monokorn mortar) to 10^{-17} m^2 for the container walls (concrete). Sorption is assumed to take place only on cementitious materials. Because the cavern liner is assumed to be fractured, a permeability of 10^{-16} m^2 is used and sorption within the liner is neglected. For the pore diffusion constant in the waste matrix and in the backfill a high value of 10^{-9} m^2/s has been chosen, whereas for the tighter materials (container walls and liner) a value of 10^{-10} m^2/s has been adopted.

For the host rock, a mean absolute permeability of 10^{-18} m^2 and a porosity of 1% is chosen. The permeability of the excavation damaged zone is assumed to be two orders of magnitude higher than the value for the intact host rock.

Table 1: Selected parameters for radionuclide transport calculations

Hydraulic Unit	**Porosity** [-]	**Absolute Permeability** [m^2]	**Pore Diffusion Constant** [m^2/s]
Waste Matrix	0.25	5e-16	1e-09
Container Backfill	0.35	1e-15	1e-09
Container Wall	0.05	1e-17	1e-10
Container Lid	0.35	1e-15	1e-09
Cavern Backfill	0.35	1e-15	1e-09
Cavern Liner	0.02	1e-16	1e-10
Excavation damaged Zone	0.05	1e-16	2e-10
Host Rock (Marl)	0.01	1e-18	2e-10
Hydraulic Gradient (regional scale): 0.4 m/m			

Hydrodynamic dispersion cannot be modelled with TOUGH2/EOS7R[1]. For compatibility, longitudinal and transversal dispersion lengths for the SEFTRAN model calculations have been set to the minimum value allowed by this code (i.e. 0.01 m).

For the modeling of the release of non-volatile radionuclides under two-phase conditions, a gas generation rate[2] of 0.5 m^3 H_2 (STP) / (y $\cdot m^3$ waste) and a very small value for the Henry constant were used. The cavern was conservatively assumed to be fully water saturated at the onset of gas generation. A Brooks-Corey type of constitutive relationship for the relative permeability was used in all hydraulic units. The residual saturations of water and gas were chosen to be 25% and 5%, respectively. Capillary pressure and compressibility of the solid phase are neglected.

In the model a hydraulic gradient of 0.4 m/m from left to right is assumed by imposing constant heads on the vertical model boundaries (Figure 2). For the transport simulations, zero concentration was prescribed on the vertical boundaries. Both for flow and transport calculations, no-flow boundary conditions were specified on the top and bottom of the model domain.

As a quantitative measure of contaminant release from the caverns to the surrounding host rock, the release rate of non-decaying species normalised to their initital mass is calculated for different distribution coefficients (K_d). This quantity is calculated at the interface between cavern liner and excavation damaged zone and is termed „fractional release rate".

Results

The comparison of the results calculated by SEFTRAN and TOUGH2/EOS7R for saturated conditions is carried out by inspection of

- stationary flow fields
- fractional release rates of sorbing, non-decaying species

The stationary flow fields are nearly identical, as expected. In Figure 3 the fractional release rates for saturated conditions are plotted as a function of time for different distribution coefficients (K_d = 0, 10^{-3}, 10^{-2}, 10^{-1}, 1 m^3/kg). For most of the relevant time period the fractional release curves corresponding to specific K_d-value are nearly identical. At early times, a minor discrepancy is observed.

In the case of the code SEFTRAN, potential grid effects are tested by using a finer mesh with 14'672 instead of 4'320 elements. This grid refinement shows no effect on the fractional release curve for K_d = 0 m^3/kg. For K_d = 10^{-2}, 10^{-1}, and 1 m^3/kg, the curves match very well for most of the time period considered. At early times, the fractional release rates calculated by the coarse mesh are slightly higher compared to the values calculated with a finer mesh.

Alternative weighting schemes for mobilities and permeabilities were investigated with TOUGH2/EOS7R by setting the parameter MOP(11) to 0 (base case), 1, and 2. It turned out that the agreement between SEFTRAN and TOUGH2/EOS7R results is best, when an

[1] Hydrodynamic dispersion can be treated with the code T2DMR which is a descendant of the TOUGH2/MULKOM family of codes (Oldenburg & Pruess 1996a)

[2] This gas generation rate was multiplied by the molecular weight ratio of air and hydrogen, in order to produce the correct molar amount of gas in the cavern.

upstream-weighting of both mobilities and permeabilities is chosen (base case). In the other cases, a significant discrepancy between SEFTRAN and TOUGH2/EOS7R results is observed at later times.

In the case with consideration of gas generation in the repository caverns, preliminary calculations for non-volatile, non-decaying species have been performed with TOUGH2/EOS7R. The calculated fractional release rates are shown in Figure 4 as a function of time and for distribution coefficients of 0 and 10^{-3} m^3/kg. The CPU-times needed for these simulations on a DEC/ALPHA 4100 (400 Mhz) are in the order 10 h, which is substantially more than the CPU-times for the calculational cases assuming saturated conditions. A comparison of Figures 3 and 4 indicates that the release of non-volatile radionuclides under two-phase conditions takes place somewhat earlier and with considerably higher rates compared to the case of fully saturated conditions. The fractional release rates are compared to the time dependent mass release of water and gas (Figure 4). This comparison shows that the release of non-volatile, non-sorbing species appears slightly retarded when compared with the release of pure water out of the caverns. This retardation is due to the travel time of dissolved species through the container walls/lid and the cavern backfill and liner. The onset of gas release takes place when a free gas path has been established from the waste through the backfill and the liner into the host rock. As can be seen in Figure 4, gas release occurs 10 years after the start of gas generation.

In this simulation, it was conservatively assumed that the caverns are fully saturated when the gas generation starts. For partially saturated conditions in the cavern at the onset of gas generation, the release of dissolved species is expected to take place at later times and with lower release rates.

Conclusions

The preliminary results of the investigation can be summarized as follows: i) The comparison of radionuclide release rates under fully saturated conditions calculated by TOUGH2/EOS7R and the standard NAGRA near field code SEFTRAN shows good agreement, and ii) the release of non-volatile radionuclides under two-phase conditions takes place somewhat earlier and with considerably higher rates compared to the case of fully saturated conditions.

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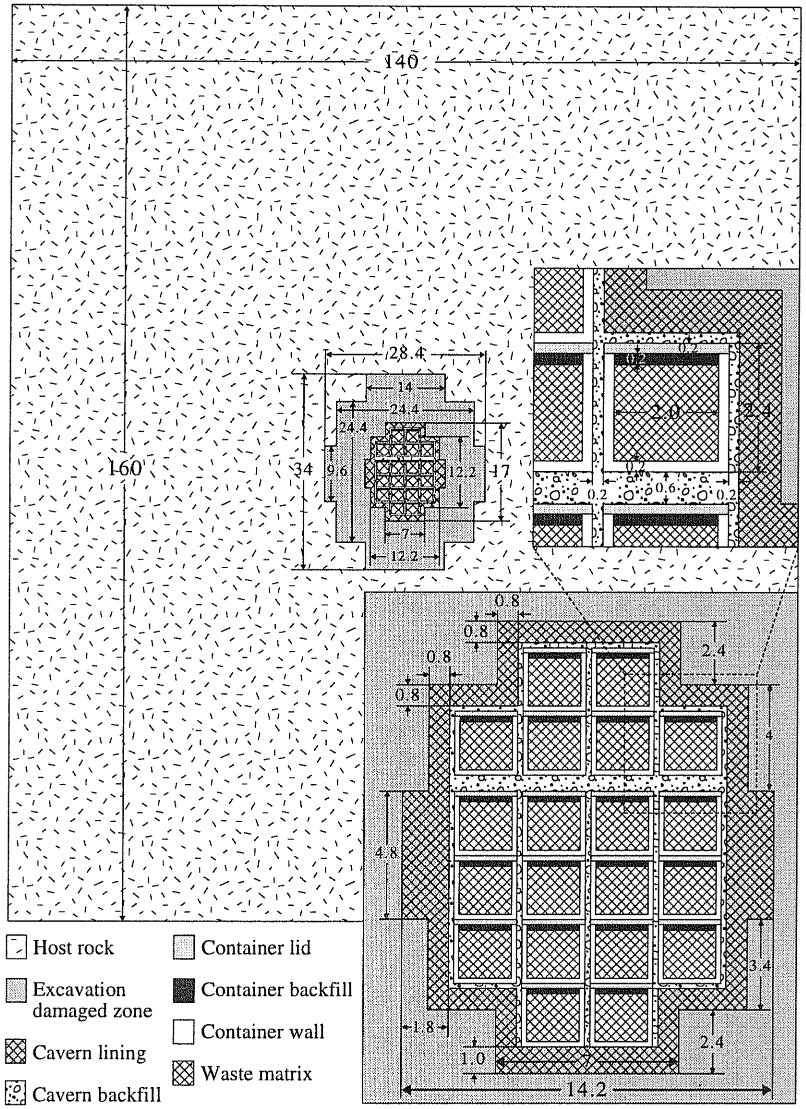

Figure 1: Model domain and definition of hydraulic units of the L/ILW repository cavern

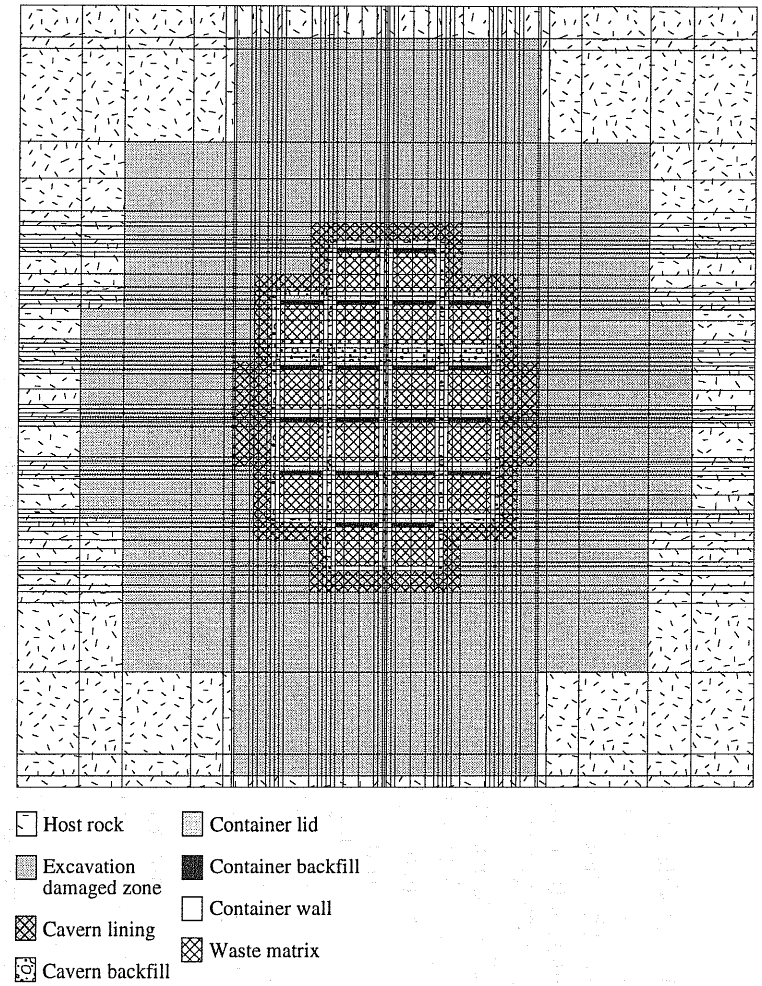

Figure 2: Finite element mesh for model simulations with SEFTRAN

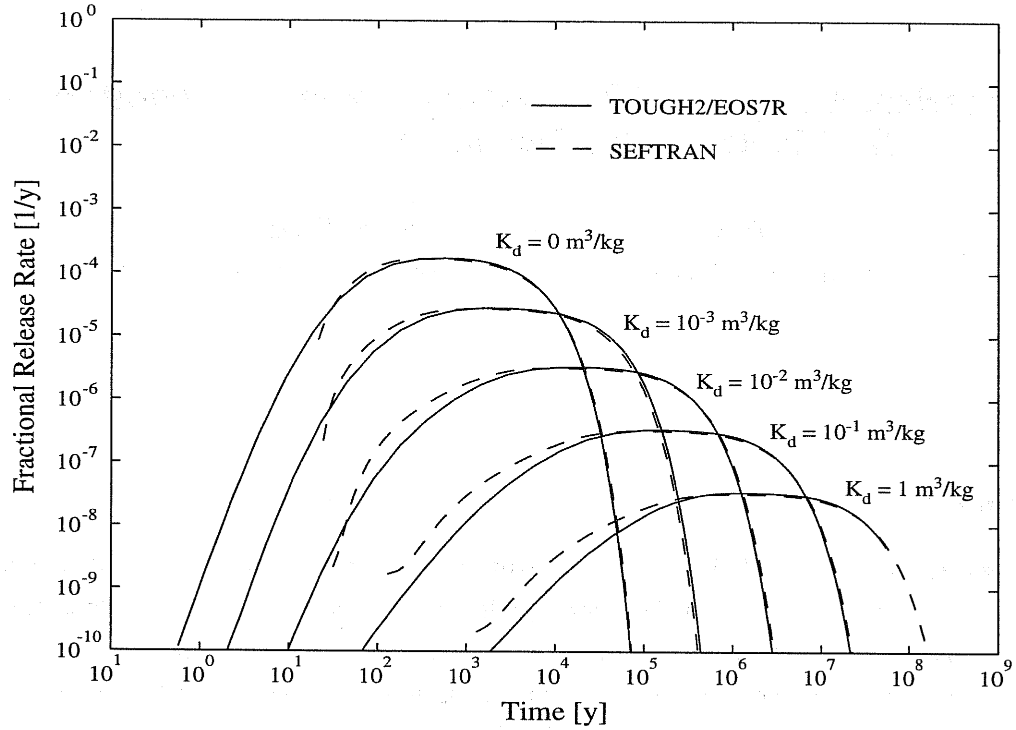

Figure 3: Fractional release rates for non-decaying species under saturated conditions as a function of time for different distribution coefficients

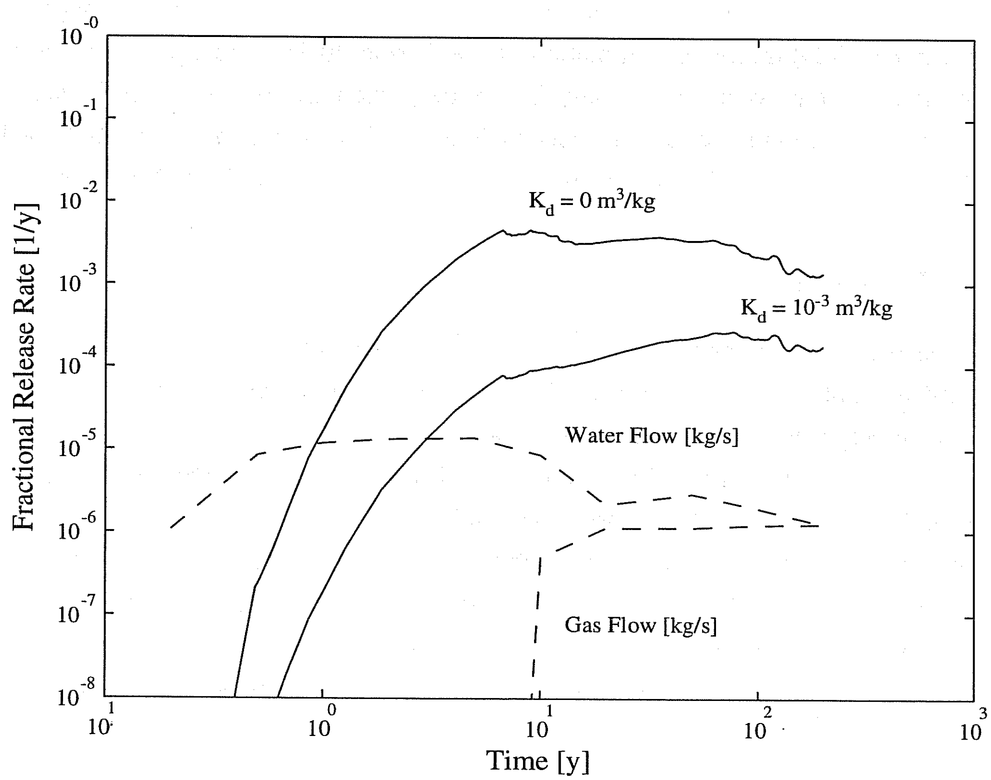

Figure 4: Fractional release rates for non-volatile, non-decaying species as a function of time for different distribution coefficients, with a gas generation rate of 0.5 m^3 H_2 (STP) / (y $\cdot m^3$ waste); also shown are water and gas release rates from the liner into the excavated disturbed zone (in kg/s)