Influence of chemical reactions on the flow system and contaminant transport in a former salt mine

St. Wilhelm[1], J. Poppei[1] , G. Mayer[1], R. Schwarz[1], G. Klubertanz[1], P. Siegel[1], B. Förster[2]

[1]Colenco Power Engineering AG, Groundwater Protection and Waste Disposal,
Mellingerstrasse 207, CH-5405 Baden, Switzerland
[2]GSF-Forschungszentrum für Gesundheit und Umwelt, Forschungsbergwerk Asse,
Am Wald 2, D-38319 Remlingen, Germany

Abstract. We have investigated the influence of large-scale chemical reactions like gas production and host rock dissolution on driving forces for fluid flow and contaminant transport in an underground waste repository located in a former salt mine. Numerical simulations were conducted to study the influence particularly of these two chemical effects. The tool of choice was the newly developed KAFKA, a code considering multi-complex physical and chemical processes of two-phase flow and nuclide transport. The sensitivity of the chemical reactions was illustrated with a series of simulation scenarios. The paper describes the modelling concept, the processes involved and – for exemplifying scenarios – the influence of gas production and host rock dissolution on the potential release of nuclides into the biosphere.

Introduction

For post-operational phase scenarios of underground radioactive waste disposals in ancient salt mines it is assumed that formation waters of different origin may penetrate the repository. This would cause mobilization of the radionuclides and, potentially, release from the repository into the biosphere.

Safety analysis and long-term prognoses rely on models. These models have to reflect the fluid flow and transport processes with consideration of various physical and chemical processes such as convergence, advection, diffusion, dispersion, dissolution/precipitation or sorption of contaminants, as well as radioactive decay. If some of these processes are coupled, the models become increasingly complex.

The main focus of this paper is on the effect of two large-scale chemical reactions: gas production and host rock dissolution. These processes have a major impact on fluid flow by altering physical properties and the geometry of the domain.

Additional complexity results from the fact that old mines are generally geometrically very complex. It is, therefore, necessary to simplify the geometry in order to achieve a reasonably simple (in terms of geometry) model (Fig.1).

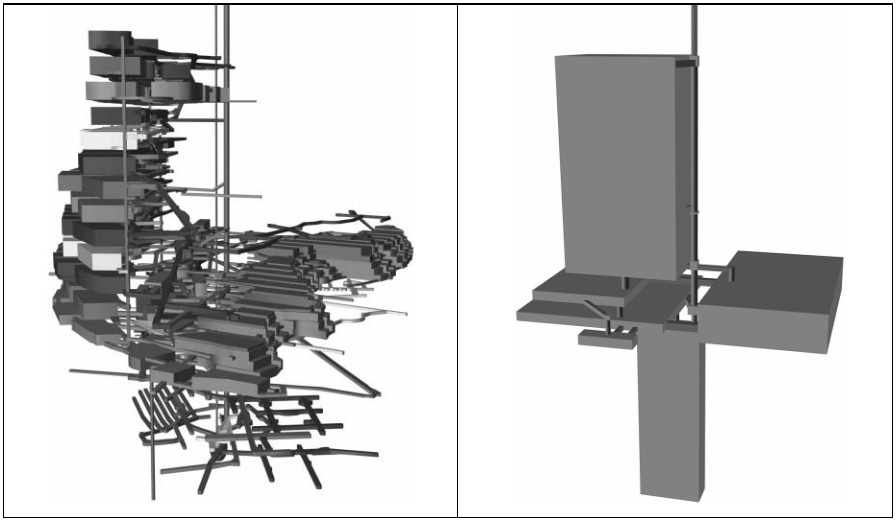

Fig. 1. Example of an actual, complex mine building (left picture) and the inferred "skeleton model"

In the following section we discuss the basic concepts of our newly developed numerical tool KAFKA = Kompartimentmodell für die Ausbreitung und die Fluiddynamik in einer konvergierenden Untertageanlage für Abfälle = Compartment model for fluid flow and contaminant transport in converging underground waste repositories (Colenco 2001). The intention is not to demonstrate the mathematical foundations of KAFKA, but to give an overview of the processes integrated.

Modelling the flow and transport system

The following processes and couplings are taken into account:

- Two-phase flow
- Diffusion and dispersion
- Sorption and limited solubility of radionuclides
- Radioactive decay
- Time-dependent gas generation due to corrosion and microbial degradation

- Dependency of pore volume (porosity) on convergence and creeping
- Dependency of flow and transport processes on varying permeability and cross-sections
- Dependency of pore volume on dissolution and precipitation
- Dependency of pressure on the chemical composition of the fluid.

The numerical solution is based on a finite volume approximation. For each finite volume, the equation of mass balance is solved for all constituents (e.g. nuclides, water, gas, salts subjected to dissolution). The processes described are (1) the volume change due to convergence and dissolution, and (2) the mass changes due to transport and chemical reactions. These changes impose alterations in dependent parameters, e.g. fluid pressure and density, which drive flow and transport.

Two-phase flow is described by the Brooks-Corey model and a generalised Darcy's law is used. The account of volume changes caused by convergence is based on the formulations in PSE 1985.

The discretisation of the domain into finite volumes implies a geometric abstraction of the mine's structure. In our concept we summarise the complex geometry of the excavated structures into four principal structural elements. These are (1) caverns, i.e. backfilled or open excavations with a high hydraulic conductivity, (2) tunnels, i.e. man-made horizontal hydraulic connections with a distinct hydraulic conductivity and which may serve as hydraulic barriers, (3) shafts, i.e. man-made vertical hydraulic connections with a distinct hydraulic conductivity, and (4) teeters, i.e. vertical connections with a hydraulic conductivity caused by stress conditions in the rock. Those structural elements are attributed specific properties with respect to hydraulic and chemical behaviour. Caverns (type 1) are, for example, treated as perfect mixing tanks while tunnels (type 2) portray conventional two-phase flow behaviour. The result is a "skeleton model" of the underground mine structures (Fig.1). Obviously, expert knowledge is essential in collapsing sets of actual mine structures into schematic model elements.

Chemical processes

The behaviour of the repository with respect to the release of radionuclides is influenced by gas generation and by the dissolution/precipitation of salts in contact with aqueous solutions.

Of the two large-scale chemical processes, the build-up of the gas pressure in the mine is obviously a direct result of chemical processes such as anaerobic corrosion of metal inventory and microbial degradation of organic matter. Based on the type of waste and the chemical environment, the effective gas generation rates as a function of time can be calculated with a separate chemical simulation model. A deduced parameterization of the temporally dependent, effective gas generation rates is then used as input for KAFKA.

In a similar manner, the dissolution and precipitation of salt (here referred to as fluid-salt interactions) in contact with aqueous solutions has been investigated in detail with an independent geochemical code, i.e. The Geochemist's Workbench

(Bethke 1998). In the system considered here, a NaCl-saturated solution is assumed to enter the mine and react with Mg-bearing salts such as carnallitite. This process results in a) a loss of carnallitite volume, b) the generation of additional solution by liberating crystal-bound water and c) the formation of a loose conglomerate of secondary minerals. Thus the fluid-salt interactions are expected to have an influence on the convergence and flow processes, as well. In KAFKA, the Mg content of the solution is used as a measure of the system's reactivity.

Model description

We chose a simple geometric model to demonstrate possible influences of gas formation and carnallitite dissolution (Fig.2). A cavern (Cav1) containing the nuclide inventory and the gas source is connected to a second cavern (Cav2) containing carnallitite. Cavern Cav2 is connected to a vertical shaft with a backfilled tunnel (Tun).

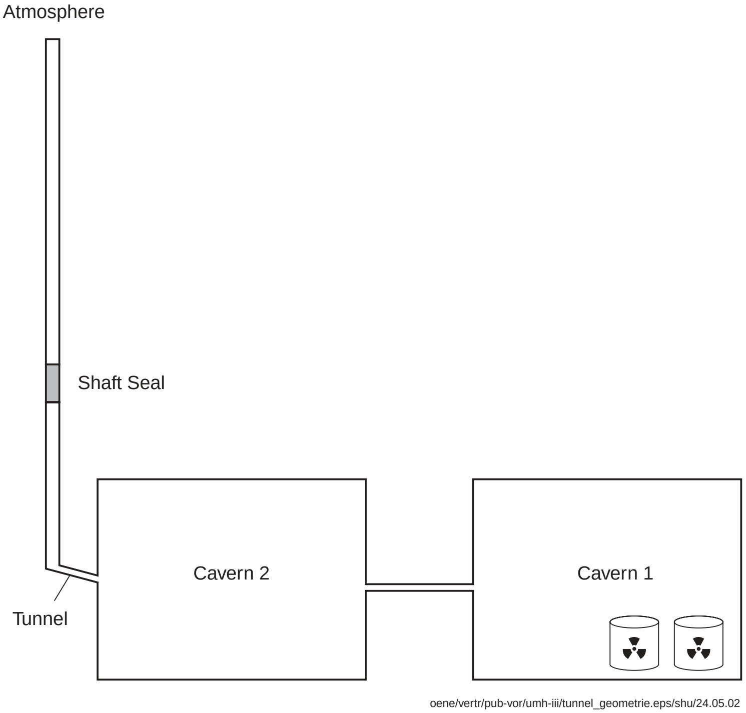

Fig. 2. Geometric configuration of the model

The caverns, the horizontal connectors between them and the tunnel are subject to convergence, whereas the shaft itself is assumed to be stable. Part of the shaft is plugged with 50 m of filling and closing materials with a permeability of 10^{-13} m^2. The tunnel (Tun) is filled with sorbing backfill materials and has a permeability of 10^{-13} m^2. The connector between the caverns is given a permeability of 10^{-11} m^2.

The diffusion is described by an overall pore diffusion coefficient of 10^{-10} m^2/s. The dispersion length generally applied is 10 m.

The waste materials in Cav1 are mobilized instantaneously. The gas production follows an empirical exponential relationship described by the gas production rate and the final total gas volume:

$$V_g(t) = V_{gk}\left(1 - e^{-\lambda_{gk} t}\right) \qquad (1)$$

with

V_{gk} ... total gas volume at infinity (m^3)

λ_{gk} ... production rate (1/s).

NaCl-solute enters at a constant rate through the top of the shaft and infiltrates into all structural elements of the model.

A simplified decay chain (N+2) and iodine (^{129}I) are used to demonstrate the influence on radionuclide concentrations under the consideration of solubility, sorption and decay. The inventory is assumed to contain the following nuclides, each with a general initial activity of 10^{10}Bq: Am^{242}, Cm^{246}, Pu^{238}, Pu^{242}, U^{234}, U^{238}, Ra^{226} and I^{129}. Both radioactive decay as well as ingrowths are taken into account.

The solubility limits and the K_D-values of the elements (the latter are required for the backfill materials in the tunnel) are listed in Table 1.

Table 1. Solubility limits and retardation coefficients used for the simulations

	Solubility limit (mol/m^3)	K_D (m^3/kg)
Am	$1.0 \cdot 10^{-3}$	0.7
Cm	$1.0 \cdot 10^{-3}$	0.7
Pu	$1.0 \cdot 10^{-3}$	0.7
U	$1.0 \cdot 10^{-3}$	$1.4 \cdot 10^{-2}$
I	$1.0 \cdot 10^{40}$	0
Th	$1.0 \cdot 10^{-3}$	0.7
Ra	$1.0 \cdot 10^{-4}$	$2.1 \cdot 10^{-3}$

The parameters outlined in this section are used for the computational cases introduced in the following sections.

Since the transport effects of sorption, diffusion, dispersion and precipitation/dissolution are not the focus of this paper, their influence on the nuclide concentration is not discussed.

Influence of chemical reactions

The sensitivity of chemical reactions was investigated with a series of simulation scenarios.

1. Influence of fluid-salt interaction only, i.e. no gas generation:

- "reference": no interaction to represent the reference case
- "half carn": interaction is limited to half of the total amount of carnallitite mass in Cav2
- "total carn": unlimited interaction with the total amount of carnallitite mass, i.e. $3 \cdot 10^8$kg.

The dissolution of carnallitite results not only in an increased pore volume (Fig.3), but also in an increase of solute out flux from the shaft into the atmosphere. The latter is due to the increase in the amount of fluid available and an increase in the volume exposed to converge (Fig.4).

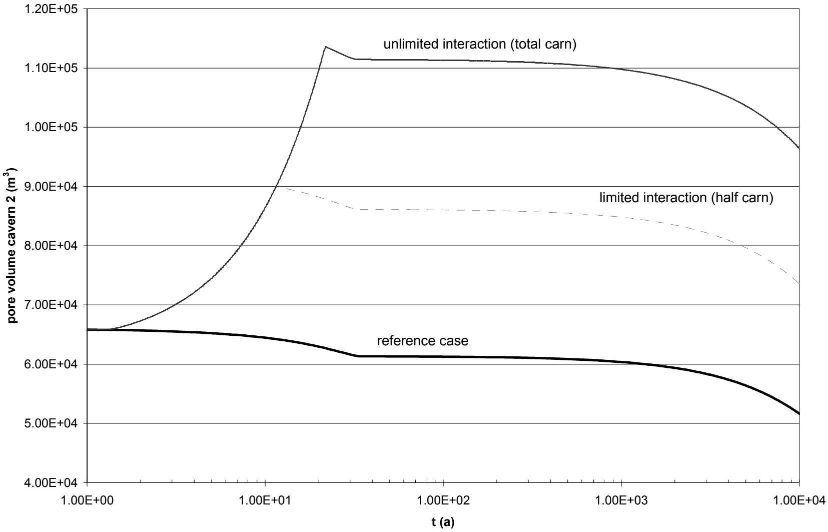

Fig. 3. Influence of carnallitite dissolution on the pore volume of the carnallitite bearing cavern Cav2

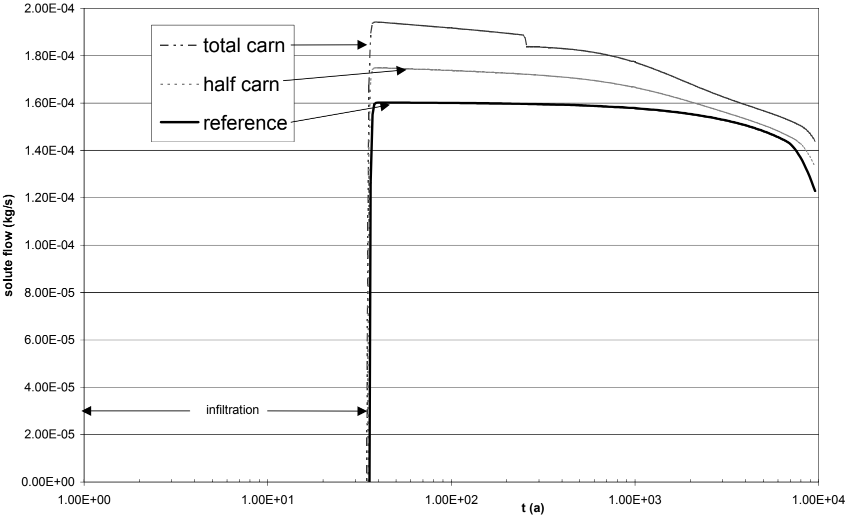

Fig. 4. Resulting solute outflux at the top of the shaft

The nuclide release into the atmosphere at the top of the shaft is element-specific as it is dependent on sorption and solubility limits (despite diffusion and dispersion). Already after about 10,000 years, the release rate for the non-sorbing iodine nuclides reaches a plateau, i.e. an equilibrium. The release rate for uranium, however, shows a marked lag (Fig.5). The release rates shown in Fig.5 are related to the initial nuclide concentrations in the inventory.

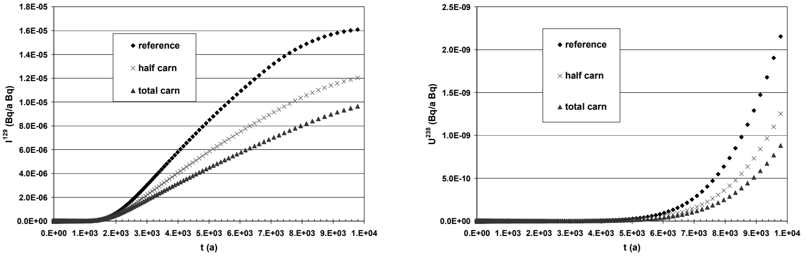

Fig. 5. Resulting annual nuclide release rate at the top of the shaft (related to the initial inventory) for iodine and uranium

It may be concluded that the fluid-salt interaction generally has an effect of retardation on the release of nuclides into the atmosphere.

2) Influence of the gas production only, i.e. no fluid-salt interaction:

- “reference”: no gas production to represent the reference case
- “slow gas”: reduced gas production rate with $\lambda_{gk}=10^{-11}$ 1/s and $V_{gk}=5\cdot10^5$ m^3
- “fast gas”: increased gas production rate with $\lambda_{gk}=10^{-10}$ 1/s and $V_{gk}=5\cdot10^5$ m^3

- "less gas": less gas volume produced with $V_{gk}=2.5\cdot10^{5}$ m^{3} and $\lambda_{gk}=10^{-11}$ 1/s.

The simulation results show that once the mine structures have filled with NaCl-solute, gas production enhances the flow of solute (now transporting radionuclides) back out into the atmosphere (Fig.6). A slow gas production rate prolongs the solute out flux and, thereby, the release of radionuclides. Therefore, depending on sorption conditions, especially late time releases are boosted by slow rates. After 10,000 years, the release rate of U^{238} is increased by a factor of 2.4 and that of I^{129} by a factor of 1.2 when compared to the reference case without gas production (Fig.7).

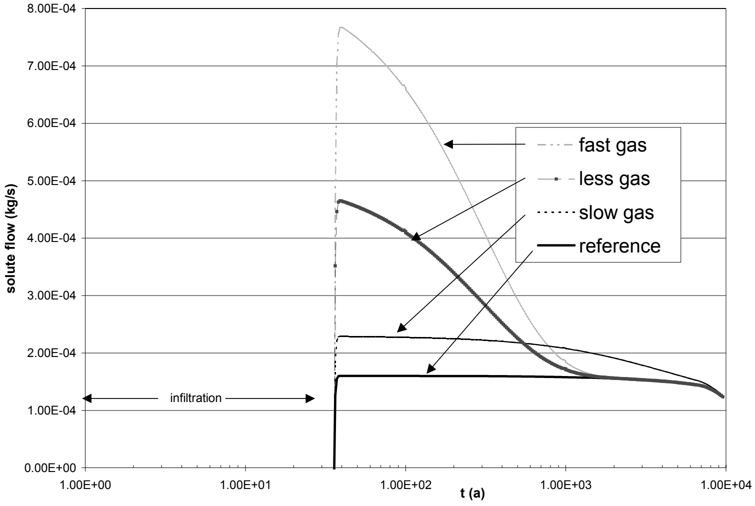

Fig. 6. Influence of gas production on solute flow through the top of the shaft

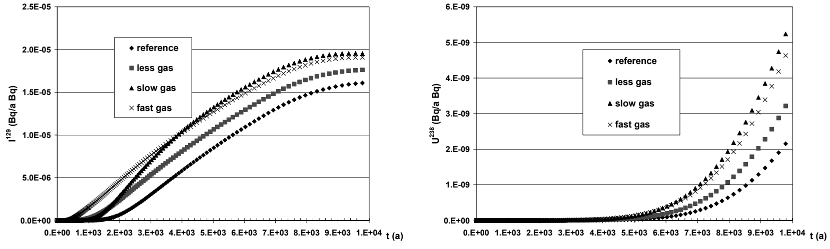

Fig. 7. Resulting annual nuclide release rate at the top of the shaft (related to the initial inventory) for iodine and uranium

3) A "realistic" scenario, i.e. with both gas generation and fluid-salt interaction:

“with gas and half carn”: interaction is limited to half of the total amount of carnallitite mass and fast gas production rate with full volume of gas produced.

Fig.8 illustrates the results as exemplified by two nuclides. The non-sorbing iodine is found to be released at a rate reduced by a factor of 0.9 (after 10,000 years) as compared to the reference case, whereas the release rate of U^{238} nuclides is increased by a factor of 1.2. At sorbing conditions (late time release) the reduction by carnallitite interaction cannot be superimposed due to gas induced release.

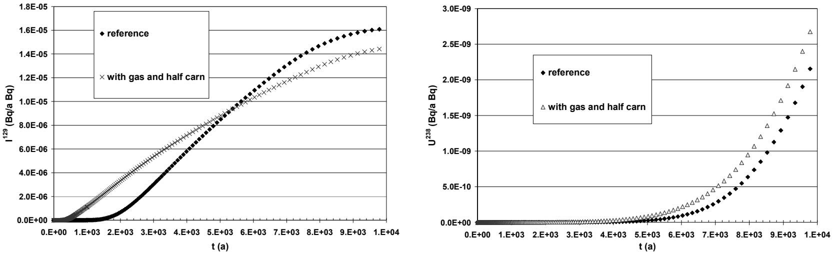

Fig. 8. Comparison of results from realistic scenarios (high gas production rate, small carnallitite amount) with reference case

Conclusions / outlook

The behaviour of the repository with respect to the release of radionuclides is influenced by gas generation and by the dissolution/precipitation of salts in contact with aqueous solutions.

The sensitivity of chemical reactions was investigated with a series of simulation scenarios.

It may be concluded that the fluid-salt interaction generally has an effect of retardation on the release of nuclides into the atmosphere. However, the effect of potential dissolution of flow barriers (containing carnallitite) is not investigated here. This would possibly result in an enhancement of outflux.

Once the mine structures have filled with NaCl-solute, the gas production enhances the flow of solute back out into the atmosphere. A slow gas production rate prolongs the solute out flux and, thereby, the release of radionuclides.

The simulation tool KAFKA has proved to provide an adequate reflection of the complexity of physical and chemical processes involved. Further simulations will consider the higher complexity of geometric conditions (e.g., Fig.1) and the treatment of the uncertainty of parameters in a probabilistic manner.

References

Bethke CM (1998) The Geochemist's Workbench®, Version 3.0. Univ. of Illinois, Urbana, 184 p.

KAFKA (2001) User’s guide, Colenco internal report

PSE (1985) Projekt Sicherheitsstudien Entsorgung (PSE): Abschlussbericht, Fachband 15: Einzeluntersuchungen zur Radionuklidfreisetzung aus einem Modellsalzstock, Berlin