



Working Report 2010-14

Modelling of Near-Field Radionuclide Transport Phenomena in a KBS-3V Type of Repository for Nuclear Waste with Goldsim Code — and Verification Against Previous Methods

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Working Reports contain information on work in progress
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The conclusions and viewpoints presented in the report
are those of author(s) and do not necessarily
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MODELLING OF NEAR-FIELD RADIONUCLIDE TRANSPORT PHENOMENA IN A KBS-3V TYPE OF REPOSITORY FOR NUCLEAR WASTE WITH GOLDSIM CODE - AND VERIFICATION AGAINST PREVIOUS METHODS

ABSTRACT

Traditional radionuclide transport models overestimate significantly some phenomena, or completely ignore them. This motivates the development of new more precise models. As a result, this work is a description of commissioning of a new KBS-3V near-field radionuclide transport model, which has been done with a commercial software called GoldSim. According to earlier models, GoldSim model uses rz coordinates, but the solubilities of radionuclides have been treated more precisely.

To begin with, the physical phenomena concerning near-field transport have been introduced according to GoldSim way of thinking. Also, the computational methods of GoldSim have been introduced and compared to methods used earlier. The actual verification of GoldSim model has been carried out by comparing the GoldSim results from simple cases to the corresponding results obtained with REPCOM, a software developed by VTT and used in several safety assessments. The results agree well.

Finally, a few complicated cases were studied. In these cases, the REPCOM's limitations in handling of some phenomena become evident. The differences in the results are caused especially by the extension of the solubility limit to the whole computational domain, and the element-wise treatment of the solubilities which was used instead of nuclide-wise treatment.

This work has been carried out as a special assignment to the former laboratory of Advanced Energy Systems in Helsinki University of Technology. The work was done at VTT.

RADIONUKLIDIEN LÄHIALUEEN KULKEUTUMISILMIÖIDEN MALLINNUS KBS-3V LOPPUSIJOTUSTILASSA GOLDSIM MALLILLA - JA VERIFIOINTI AIKAISEMPIIN MENETELMIIN VERRATEN

TIIVISTELMÄ

Perinteiset radionuklidien kulkeutumismallit yliarvioivat reilusti joitakin ilmiöitä tai jättävät ne kokonaan huomiotta. Tämä motivoi uusien tarkempien mallien kehittämistä. Tässä työssä kuvataan uuden GoldSim ohjelmalla tehdyn KBS-3V konseptin mukaisen radionuklidien lähialuekulkeutumismallin käyttöönotto. Aikaisempien mallien tapaan GoldSim mallissa käytetään rz -koordinaatistoa, mutta radionuklidien liukoisuusrajoitteisuus on käsitelty aiempaa tarkemmin.

Aluksi radionuklidien kulkeutumiseen liittyvät ilmiöt on kuvattu GoldSimin ajatusmallin mukaisesti, laskennalliset menetelmät on esitelty ja näitä on verrattu aiemmin käytettyihin menetelmiin. Varsinainen GoldSim mallin verifiointi on suoritettu vertaamalla yksinkertaistetuista tilanteista saatuja tuloksia REPCOM-ohjelmalla saatuihin vastaaviin tuloksiin. REPCOM on VTT:n kehittämä useissa turvallisuusanalyysissä käytetty ohjelma. Tulokset vastaavat toisiaan hyvin.

Lopuksi yksinkertaisista tilanteista on siirrytty mutkikkaampiin, joissa perinteisen REPCOM mallin rajat ilmiöiden käsittelyssä tulevat vastaan. Eroja tuloksiin aiheuttavat erityisesti uudessa mallissa tehtävä liukoisuusrajojen ulottaminen koko laskenta-alueelle ja rajojen käsitteleminen alkuainekohtaisesti nuklidikohtaisen käsittelyn sijaan.

Tämä työ on tehty erikoistyönä Teknillisen korkeakoulun entiseen Energiatieteiden laboratorioon. Työn tekopaikkana toimi VTT.

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1 INTRODUCTION

Nuclear energy has been powering the Finnish industry and households over three decades. During this time, spent fuel has accumulated in the intermediate storages nearby the power plants. The Finnish final disposal plan for spent fuel is, in brief, to seal the fuel rods into iron-copper canisters, surround the canisters with bentonite, a clay-like well-expanding, diffusion-dominating material, and place the whole package approximately half kilometer deep into the bedrock. The idea sounds simple but the planning and the execution are quite demanding. Due to the long-lasting radioactive waste material and the public image of nuclear power, the research on disposal must be thorough and extensive as well as intelligible to authorities and public. The licensing of such a geologic repository for spent fuel requires, among other things, a safety case, that is, a summary of all arguments that show that there is reasonable assurance that the spent fuel management solution remains safe for a long time.

The regulatory requirements for a spent fuel repository at Olkiluoto are set out in the Government Decision on the safety of the disposal of spent nuclear fuel (STUK, 1999) and, in more detail, in Guide YVL 8.4 issued by the Finnish regulator STUK (2001). These requirements are, however, currently under revision. Guide YVL 8.4 distinguishes between the "environmentally predictable future" (also referred to by the regulator as "several thousand years"), during which conservative estimates of dose must be made (i.e. estimates that tend to over-estimate dose where there is uncertainty), and the "era of large-scale climate changes" when periods of permafrost and glaciations are expected, and radiation protection criteria are based on constraints on nuclide-specific activity fluxes from the geosphere, termed "geo-bio flux constraints". YVL 8.4 also gives a qualitative requirement that: "The barriers shall effectively hinder the release of disposed radioactive substances into the host rock for several thousands of years."

This report focuses on the radionuclide release and transport model in a case of an early canister failure, which is very unlikely.

2 BACKGROUND

2.1 Actors in Nuclear Waste Management

To understand the context and the needs the modelling is facing, a brief introduction to the circumstances of nuclear waste management in Finland is needed. In Finland, the spent fuel disposal is managed by Posiva Oy. It is owned and funded by the energy corporations Fortum Oy and Teollisuuden Voima Oy that produce nuclear energy. Posiva carries out research on its own or contracts it out to external organisations, such as VTT, Technical Research Centre of Finland. The third party on the disposal field is STUK, the Radiation and Nuclear Safety Authority. It regulates the safety of handling, storage and disposal of nuclear waste. The Ministry of Employment and the Economy handles the general regulation of the final disposal, decides on principle and establishes the time schedules to be implemented by the power companies. A schematic presentation of the counterparties is shown in Figure 2-1.

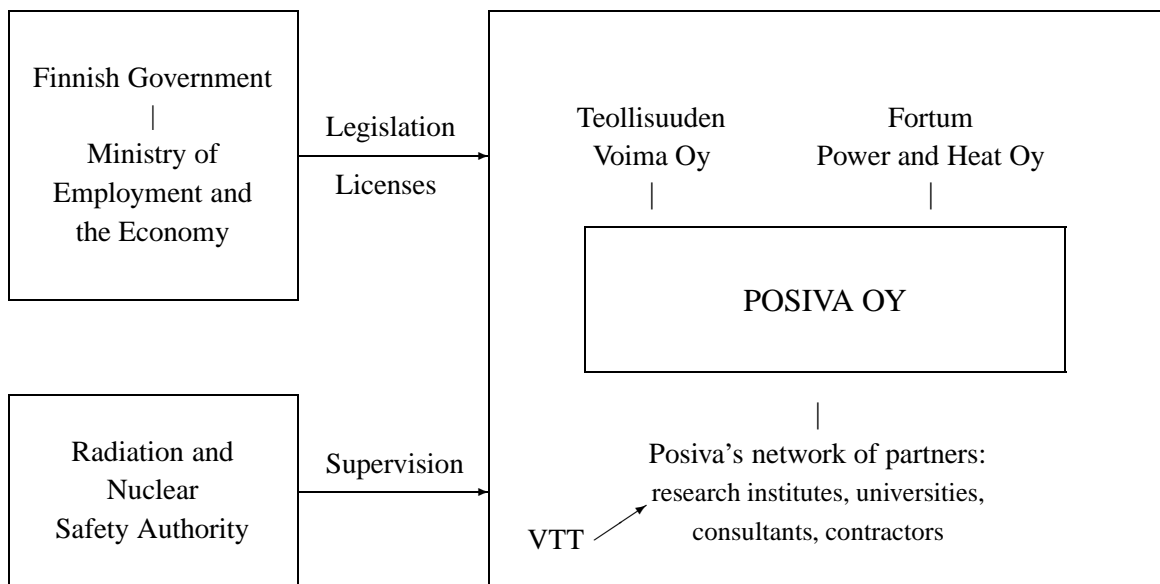


Figure 2-1. The main Finnish nuclear waste management actors and their relations. The picture is a replica of a picture at Posiva's homepage (POSIVA, 2008) with additional VTT text.

2.2 The State of the Finnish Disposal Plan

The Finnish disposal plan, touched briefly in the introduction, is presented in schematic pictures in Figure 2-2. Due to the political consensus, the Finnish plan has evolved into

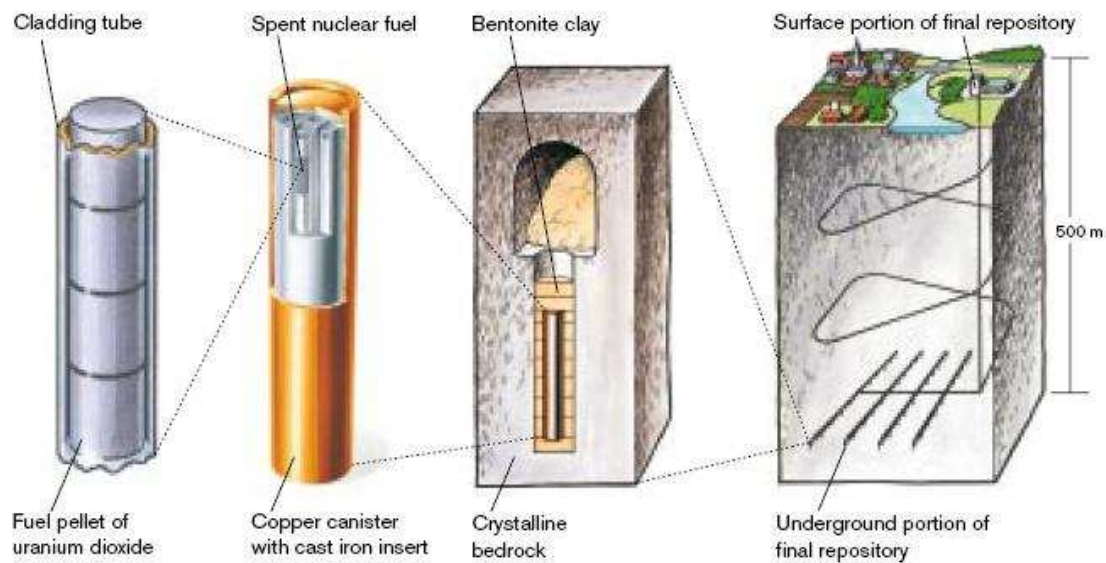


Figure 2-2. A picture series of the disposal site. The Finnish plan follows essentially the Swedish KBS-3V plan and the picture series is from a Swedish technical report (SKB, 2006b).

quite a mature state. The next grand step is to apply a construction license to dispose the nuclear waste. Posiva plans to submit the application to STUK in 2012. Not surprisingly, the planning and the research of the disposal is speeding up. At the same time, new modelling tools arise and computational capabilities improve. As a result, more realistic and developed models are wanted. However, the models have to be kept simple enough so that they can be interpreted clearly. To meet these challenges in the field of radionuclide transport, VTT has introduced a new simulation program called GoldSim, the main object of this report.

2.3 Radionuclide Transport Computation at VTT

The principle of conservativeness is used when computing the release of the disposed radionuclides. It means that all simplifying estimates should be made in a way that they do not underestimate the effects of the release.

At VTT, the radionuclide transport from the repository to biosphere has mostly been computed with a in-house code called REPCOM, REPository COMpartments. It has been programmed with `fortran77` and is rather old, but it works fine, especially with modern computers. However, it makes some overly conservative estimates that mask some significant phenomena. The present-day tendency to make the estimates as real as possible without losing the conservativeness is not really followed. One more obstacle

preventing the widespread usage of REPCOM is the unclarity of the user interface.

Some tests have been made with a program called PORFLOW to confirm the calculations made with REPCOM. The results obtained with the programs coincide well and have been published in the technical report (Nordman and Vieno, 2003).

For further information about the radionuclide transport computing at VTT, contact REPCOM specialist Lic.Sc. Henrik Nordman or get acquainted with the technical reports (Nordman and Vieno, 2003), (Nordman and Vieno, 1994), and (Vieno and Nordman, 2000).

3 PHYSICAL PHENOMENA

3.1 Sorption and Retardation in a Porous Medium

Porous materials tend to react with nuclides by sorbing them on surfaces. The degree of sorption is described by the distribution coefficient¹ K_d , which is defined as

$$K_d = \frac{m_s/M_s}{m_f/V_f} \quad (1)$$

where

K_d	=	the distribution coefficient	(m ³ /kg)
m_s	=	the mass of the nuclide sorbed on the solid material	(kg)
M_s	=	the mass of the solid material	(kg)
m_f	=	the mass of the nuclide in the fluid (typically water)	(kg)
V_f	=	the volume of the fluid	(m ³).

Partition coefficient can also be defined between two fluids or two solids. If the volumes of fluids and the masses of solids are denoted with quantities U , Equation (1) becomes

$$K_{1,2} = \frac{m_1/U_1}{m_2/U_2} \quad (2)$$

$K_{1,2}$	=	the distribution coefficient between materials 1 and 2	(m ³ /kg or kg/m ³ or -)
m_i	=	the mass of the nuclide in material i	(kg)
U_i	=	the quantity of the material i , m for solids and V for fluids	(kg or m ³).

For computational purposes, it is useful to define a reference fluid which all the other materials are compared to. This is what GoldSim does and, thus, the following notation is adopted from the GoldSim user's guide (GoldSim, 2007). For each substance a partition coefficient $K_{material,ref}$ can be expressed vis-à-vis the reference fluid. Consequently, the partition coefficient between two substances can be calculated from their reference partition coefficients:

$$K_{1,2} = \frac{K_{1,ref}}{K_{2,ref}}. \quad (3)$$

In addition, the units of the reference partition coefficients have units [volume]/[mass] for solids and [volume]/[volume] for fluids. If, again, the volumes V 's and the masses

¹The distribution coefficient is also called a partition coefficient.

M 's are referred as quantities U of medium corresponding the partition coefficients, the concentration of the nuclide n in material i is

$$c_{n,i} = \frac{K_i}{\sum_j (K_j U_j)} m_n \quad (4)$$

where

$$\begin{aligned} c_{n,i} &= \text{the concentration of the nuclide } n \text{ in material } i && (\text{kg/m}^3) \\ K_i &= K_d \text{ between material } i \text{ and the reference fluid} && (\text{m}^3/\text{kg}, \text{kg/m}^3 \text{ or } -) \\ K_j &= K_d \text{ between material } j \text{ and the reference fluid} && (\text{m}^3/\text{kg}, \text{kg/m}^3 \text{ or } -) \\ U_j &= \text{the quantity of the material } j && (\text{kg or m}^3) \\ m_n &= \text{the mass of the nuclide } n \text{ in the volume} && (\text{kg}). \end{aligned}$$

The summation is taken over all the materials in the volume. In this report, all the volumes contain only water and single solid material. Thus, the concentration in water can be calculated using K_d from Equation (1) in Equation (4). However, for further use of the model the above general form of concentration used by GoldSim is introduced.

Retardation of the mass transport through a medium, caused by the sorption and the pores, is specified by the retardation factor R :

$$R = \frac{m_t}{m_f} = \frac{m_s + m_f}{m_f} \quad (5)$$

where

$$\begin{aligned} R &= \text{the retardation factor} && (-) \\ m_t &= \text{the total mass of an element} && (\text{kg}) \\ m_f &= \text{the mass of an element in the fluid} && (\text{kg}) \\ m_s &= \text{the mass of an element in the solid} && (\text{kg}). \end{aligned}$$

In a porous material, the retardation factor is

$$R = \frac{m_t}{m_f} = 1 + \frac{m_s}{m_f} = 1 + \frac{M_s K_d}{V_f} = 1 + \frac{(1 - \varepsilon) V_t \rho_s K_d}{\varepsilon V_t} = 1 + \frac{1 - \varepsilon}{\varepsilon} \rho_s K_d \quad (6)$$

where

ε	=	the porosity ² of the material	(-)
m_t	=	the total mass of an element	(kg)
m_f	=	the mass of an element in the fluid	(kg)
m_s	=	the mass of an element in the solid	(kg)
M_s	=	the mass of the solid	(kg)
K_d	=	the distribution coefficient	(m ³ /kg)
V_f	=	the volume of the fluid	(m ³)
V_t	=	the total volume	(m ³)
ρ_s	=	the dry density of the solid	(kg/m ³).

(Nordman and Vieno, 1994) (GoldSim, 2007)

²Geometrically seen, porosity is the percentage of the pore volume in a material. However, in some cases all the pore volume is not available for some substances. For example, anion exclusion reduces the space for anions in clay. In these cases, the porosity should be considered rather as a diffusion related parameter than as a pure geometric property.

3.2 Diffusion through a Porous Medium

In a simple fluid, the diffusion obeys Fick's law

$$\vec{j}_f = -D_f \nabla c_f \quad (7)$$

where

$$\begin{aligned} \vec{j}_f &= \text{the mass flux}^3 \text{ of the substance in a fluid } f && (\text{kg}/(\text{m}^2 \cdot \text{s})) \\ D_f &= \text{the diffusion coefficient} && (\text{m}^2/\text{s}) \\ c_f &= \text{the concentration of the substance in the fluid } f && (\text{kg}/\text{m}^3) \\ \nabla &= \text{gradient operator.} \end{aligned}$$

In a porous media, the geometry complicates the diffusion and the equation takes the form

$$\vec{j}_{porous} = \epsilon \frac{\delta}{\tau^2} \vec{j}_f = -\epsilon \frac{\delta}{\tau^2} D_f \nabla c_f = -\epsilon G D_f \nabla c_f = -D_e \nabla c_f \quad (8)$$

where

$$\begin{aligned} \vec{j}_{porous} &= \text{the mass flux in the porous media} && (\text{kg}/(\text{m}^2 \cdot \text{s})) \\ \delta &= \text{the constrictivity} && (-) \\ \tau &= \text{the tortuosity} && (-) \\ G &= \text{the factor denoting the geometric properties} && (-) \\ &\quad \text{of the pores on diffusion}^4 && (-) \\ \epsilon &= \text{the porosity} && (-) \\ D_f &= \text{the diffusion coefficient of the fluid} && (\text{m}^2/\text{s}) \\ D_e &= \text{the effective diffusion coefficient of the fluid} && (\text{m}^2/\text{s}). \\ &\quad \text{in the medium} && \end{aligned}$$

The mass flow through a surface S is obtained by integrating the previous equation over the surface, i.e.

$$\dot{m} = - \int_S D_e \nabla c_f \cdot \vec{n} dS \quad (9)$$

where \vec{n} is the normal vector of the surface.

³As distinct from molic flux presented by capital \vec{J} , the mass flux is denoted by lower case \vec{j} . Of course, in the molic flux case, also the concentration should be presented in moles.

⁴GoldSim uses tortuosity τ in the place of G . This should be noted to avoid mistakes.

3.3 Solubility Limits

When an element is dissolved in a fluid, sometimes its concentration is limited by its solubility. The limit is called the solubility limit. If the amount of the element in a volume is more than what can be dissolved, the exceeding part precipitates. The solubility limit is an element property. Thus, the isotopes of an element share the solubility in the isotopic ratio, and this complicates the mass transport computation a bit.

GoldSim uses mass transport approach, and thus the following equation is needed. Given a volume V , the amount of element saturating the fluid is m_{sat} . It can be determined by

$$m_{sat} = n^r \sum_i K_i U_i \quad (10)$$

where

m_{sat}	=	the mass of the element causing the saturation in the volume	(kg)
n^r	=	the solubility limit of the reference fluid	(mol/l)
K_i	=	K_d between material i and the reference fluid	(m ³ /kg, kg/m ³ or -)
U_i	=	the quantity of the material i , m for solids and V for fluids	(kg or m ³).

The summation is taken over all the mediums in the volume V .

3.4 Radioactive Decay

The well-known differential equation for radioactive decay for some nuclide is

$$\dot{N} = -\lambda N \quad (11)$$

where

N	=	the number of the radioactive nuclei	(-)
\dot{N}	=	the time derivative of N	(1/s)
λ	=	the decay constant	(1/s).

The half-life of a nuclide can be derived from:

$$T_{1/2} = \frac{\ln 2}{\lambda}. \quad (12)$$

Equation (11) can be written with respect to the mass of a nuclide:

$$\dot{m} = -\lambda m \quad (13)$$

where

$$\begin{aligned} m &= \text{the mass of a nuclide} && (\text{kg}) \\ \dot{m} &= \text{the time derivative of } m && (\text{kg/s}). \end{aligned}$$

When a radionuclide decays, it becomes another nuclide, called a daughter nuclide. If an overall mass change of the nuclide i due to the radioactive decay is wanted, the rate of mass change becomes

$$\dot{m}_i = -\lambda_i m_i + \sum_j f_j R_j \frac{A_i}{A_j} \lambda_j m_j \quad (14)$$

where

$$\begin{aligned} \lambda_i &= \text{the decay constant of nuclide } i && (1/\text{s}) \\ f_j &= \text{the fraction of the parent nuclide } j \text{ that decays} \\ &\quad \text{to the nuclide } i \\ R_j &= \text{the stoichiometric ratio of moles of nuclide } i \\ &\quad \text{produced per mole of nuclide } j \text{ decayed} && (1/\text{s}) \\ A_i &= \text{the atomic weight of the nuclide } i && (\text{g/mol}) \end{aligned}$$

The summation is taken over all the direct parent nuclides of the nuclide i .

If a heavy radionuclide decays to a daughter nuclide, the daughter tends to decay further. Nuclides of this nature form specific decay chains. However, for computational purposes, it is practical to define a bit larger concept, a decay family. Simply, it consists of all the decay chains that have nuclides of common elements.

3.5 Connecting the Phenomena

All the preceding mass change rates can be combined into one equation. If a volume V is considered, the mass change rate is

$$\dot{m}_V = -\lambda_i m_i + \sum_j f_j R_j \frac{A_i}{A_j} \lambda_j m_j - \int_{\partial V} D_e \nabla c_f \cdot \vec{n} dV + S \quad (15)$$

where Equations (14) and (9) have been combined, and a source term S has been added.

4 COMPUTATIONAL METHODS

This section is a description of the mass transport solving techniques that GoldSim is based on ⁵. The methods used in REPCOM are also discussed here.

4.1 The Approach to the Mass Transport Problem

GoldSim uses a control or finite volume approach to solve the mass transport problem of the radionuclides. The idea is to divide the region examined in small volumes and compute the mass changes in them. The small volumes are often called control or finite volumes, but the GoldSim uses the term cell. Consequently, the term cell is also used in this report. Further, the computational region, i.e. the cells, is called cell net. The mass of a nuclide in a cell can change by diffusion to the neighbouring cells, by radioactive decay, or by the radioactive decay of the parent nuclides. These phenomena are described by applying Equation (15) to each cell and discretizing the diffusion. Of course, the obtained equations have to be discretised also timewise. In a cell, the mixing and the sorption of the nuclides is assumed to happen instantaneously. The cell net uses cylindrical geometry without angular dependency.

4.2 Discretizing the Diffusion

When the solubility limit is not reached, the diffusive mass flow for a nuclide from a cell i to a neighbouring cell j is

$$\dot{m}_{d,i \rightarrow j} = Q_{i \rightarrow j} \left(c_i - \frac{c_j}{K_{ji}} \right) \quad (16)$$

where

c_i	=	the dissolved concentration of a nuclide in the cell i	(kg/m ³)
$Q_{i \rightarrow j}$	=	the diffusive conductance between the cells i and j	(m ³ /s)
K_{ji}	=	the distribution coefficient between the fluids in the cell j and i	(-).

The diffusive conductance of the cell i in the $i \rightarrow j$ direction with a simple discretization of the diffusion ⁶ is

$$Q_{i,i \rightarrow j} = \frac{D_{e,i} A}{\Delta x_i} \quad (17)$$

⁵For further details of the GoldSim techniques, see the GoldSim manual (GoldSim, 2007)

⁶The discretization takes only the nearest neighbours into account. In the orthogonal uniform xy -coordinates this equals central difference with truncation error $O(\Delta x^2, \Delta y^2)$ (Siikonen, 2006)

where

$$\begin{aligned}
 A &= \text{the diffusive area} && (\text{m}^2) \\
 \Delta x_i &= \text{the diffusive length in the cell } i && (\text{m}) \\
 D_{e,i} &= \text{the effective diffusion coefficient in the cell } i && (\text{m}^2/\text{s}).
 \end{aligned}$$

The diffusion conductance in the cell j has the same form. Consequently, these conductances in series are the conductance between the cells,

$$Q = \frac{A}{\frac{\Delta x_i}{D_{e,i}} + \frac{\Delta x_j}{D_{e,j}}}. \quad (18)$$

The mass flow in Equation (16) can be divided into two parts, one of which is dependent only of the mass in the cell i and drives mass from the cell i to j . Correspondingly, the other drives mass from cell j to i and depends only on the mass in cell j . In other words, the unidirectional diffusive mass flow is calculated assuming that the concentration in the other cell is zero. The sum of two opposite unidirectional flows equals the whole mass flow. According to this idea, a unidirectional diffusive flow for a nuclide $f_{i \rightarrow j}$ is introduced as

$$f_{i \rightarrow j} = t_{i \rightarrow j}^d m_i \quad (19)$$

where $t_{i \rightarrow j}^d$ is a transfer coefficient for dissolved nuclide from cell i to j specified by the diffusive conductance in Equation (18) and the distribution coefficients of the nuclide in question between materials in cells i and j .

4.3 Dealing with Nonlinearity when a Solubility Limit is Reached

When the solubility limit is reached, the unidirectional diffusive flow in Equation (19) becomes

$$f_{i \rightarrow j} = t_{i \rightarrow j}^d \frac{n_i^s}{n_i} m_i \quad (20)$$

$$\begin{aligned}
 t_{i \rightarrow j}^d &= \text{the transfer coefficient} && (1/\text{s}) \\
 m_i &= \text{the mass of the nuclide} && (\text{kg}) \\
 n_i &= \text{the total amount of the element in the cell } i && (\text{mol}) \\
 n_i^s &= \text{the solubility limit of the element in the cell } i && (\text{mol}).
 \end{aligned}$$

The equation is non-linear and to solve it, GoldSim applies the first order Taylor expansion at reference points that are chosen to meet the accuracy demands. If the values at a

reference point are denoted by an overline (\bar{m}, \bar{n}) and the cell indexes are dropped, the first order expansion of Equation (20) is

$$f \approx t^d \frac{n^s}{\bar{n}} \bar{m} + t^d n^s \sum_{iso} \left[\frac{\partial}{\partial m_{iso}} \left(\frac{m}{n} \right)_{ref} (m_{iso} - \bar{m}_{iso}) \right] \quad (21)$$

where

$t_{i \rightarrow j}^d$	=	the transfer coefficient	(1/s)
m	=	the mass of the nuclide	(kg)
n	=	the total amount of the element	(mol)
n^s	=	the solubility limit of the nuclide	(mol)
m_{iso}	=	the mass of a isotope of the same element as the nuclide that is being computed	(kg).

The summation takes all the isotopes of the element into account and the derivatives are evaluated at the reference point. The summation term can be further expanded as

$$\begin{aligned} \sum_{isotopes} \left[\frac{\partial}{\partial m_{iso}} \left(\frac{m}{n} \right)_{ref} (m_{iso} - \bar{m}_{iso}) \right] &= \frac{m_s - \bar{m}_s}{\bar{n}} - \frac{\bar{m}}{\bar{n}^2} \sum_{isotopes} \frac{m_{iso} - \bar{m}_{iso}}{M_{r,iso}} \\ &= \frac{m_s - \bar{m}_s}{\bar{n}} - \frac{\bar{m}}{\bar{n}^2} \sum_{isotopes} \frac{m_{iso}}{M_{r,iso}} + \frac{\bar{m}}{\bar{n}} \end{aligned} \quad (22)$$

where M_r denotes the molecular weight. With the summation (22) and rearranging, the original Equation (21) can be written as

$$f \approx (m + \bar{m}) t^d \frac{n}{\bar{n}} - \bar{m} t^d \frac{n}{\bar{n}^2} \sum_{isotopes} \frac{m_{iso}}{M_{r,iso}}. \quad (23)$$

GoldSim uses Equation (19) to calculate the mass transport from cell to another whenever possible. However, at some point the solubility limit is reached and the program has to switch to Equation (23) to calculate the diffusion. The problem is when to change the equation. If the time-step is too long, the solubility limit is exceeded which is physically not realistic. To avoid the unrealistic result, GoldSim uses fractional time-stepping when the limit is approached. It shortens the time-steps so that the change is made time-wise reasonably precisely. With high precision settings the maximum solubility overshoot is 2.5 per cent, and the shortest time-step is 10^{-8} th of the user specified step. For further information about the precision settings, see appendix A.

4.4 The Cell Net Solution Algorithm

When the discretized unidirectional diffusive flows are applied to the whole region, the mass change Equation (15) for one nuclide can be expressed in a matrix form. However, because of the decay properties and the element nature of the solubility, a whole decay family has to be solved at once. If the masses of each nuclide in each cell are arranged into a vector m , an equation

$$\dot{m} = [D + T] m + S \quad (24)$$

is obtained. The symbols are

- m = vector containing the masses of each nuclide of a decay family in each cell
- \dot{m} = the time derivative of m
- D = the matrix which all the decay and ingrowth coefficients⁷
- T = the matrix which contains the mass transfer coefficients between the cells, i.e. the same coefficients as in the unidirectional diffusion equations (19) and (20). Also, direct flow rates can be given to the cells with the mass transfer coefficients, for example, for boundary condition purpose.
- S = the vector containing the sources of the nuclides.

GoldSim solves Equation (24) with implicit Euler method, also called first order backwards difference formula if looked from another point of view.⁸ The method applied to Equation (24) gives

$$\frac{m(t + \Delta t) - m(t)}{\Delta t} = [D + T] m(t + \Delta t) + S(t) \quad (25)$$

where Δt is a time-step. A basic form of Equation (25) is

$$[I - D\Delta t - T\Delta t] m(t + \Delta t) = m(t) + S(t)\Delta t \quad (26)$$

where I is the identity matrix. The length of the m vector for a decay family is $m \times n$ where m is the number of the nuclides and n is the number of the cells in the cell net. In consequence, the size of the matrices I , D , and T is $(m \times n) \times (m \times n)$.

⁷The ingrowth coefficients express the rate at which the parent nuclides decay to the daughters.

⁸The method is unconditionally stable and conserves mass. For further information about the method, consult any proper book on numerical solutions of partial or ordinary differential equations, for example (Smith, 1985), (Larsson and Thomée, 2003), or (Burden and Faires, 1983).

4.5 Solving the Linear Equations

Solving the sparse matrix Equation (26) is a matter of numerical linear algebra. GoldSim uses a two-step, fail-safe approach with iterative methods. Firstly, it solves the system with the Bi-Conjugate Gradient Stabilized method, BiCGS. If the method fails for any reason, Generalized Minimum RESidual method, GMRES, is applied. BiCGS is a robust and efficient method, but not as robust and general as the GMRES. However, BiCGS is preferred to GMRES as the solution method, because GMRES is somewhat slower. For further information about the linear algebra, see books (Barrett et al., 1994), (Trefeten and Bau III, 1997), and webpage (IML++, 2008).

4.6 The REPCOM Approach

This section is a quick review of the methods included in REPCOM for comparison. REPCOM uses the same kind of diffusion model as GoldSim, i.e. the radionuclides diffuse to the neighbouring compartments (cells) with a rate specified by the concentration difference, diffusive length, area, and coefficient. However, REPCOM does not take the solubility limits into account outside the disposal canister. Also, the decay chains are computed separately. Consequently, the solubility is treated as a nuclide-specific property, even though, strictly speaking, it is an element-specific property. These nonrealistic simplifications may lead to too fast release rates, and so, they are the main reasons to change REPCOM to GoldSim as a modelling tool. With the simplifications, the equation to be solved takes the form

$$\frac{d\mathbf{A}}{dt} = \mathbf{B} \mathbf{A}(t) + \mathbf{S}(t) \quad (27)$$

where

- \mathbf{A} = the the vector containing the nuclide inventories in the compartments
- \mathbf{B} = the diffusive coefficient matrix
- \mathbf{S} = the vector containing the nuclide sources.

REPCOM solves this equation in an unusual manner. It uses the well-known analytical solution

$$\mathbf{A}(t) = e^{\mathbf{B} \cdot (t-t_0)} \mathbf{A}(t_0) + \int_{t_0}^t e^{\mathbf{B} \cdot (t-\tau)} \mathbf{S}(\tau) d\tau \quad (28)$$

to the ordinary differential equation, Eq. (27), and calculates the matrix powers with the eigenvalue decomposition. This is contrary to conventional methods that solve Equation

(27) directly. The eigenvalues and -vectors are calculated with LAPACK's `dgeev`⁹ subroutine. The computational cost of finding all the eigenpairs of a large matrix is considerable compared to solving an equal size linear equation. In consequence, the method based on the eigenvalue decomposition is not recommended for large problems.

In principle, the exponential of a matrix could be computed in many ways. Methods involving approximation theory, differential equations, the matrix eigenvalues, and the matrix characteristic polynomial have been proposed. In practice, consideration of computational stability and efficiency indicates that some of the methods are preferable to others, but that none are completely satisfactory (Moller and Van Loan, 1978). In a repository for a nuclear waste the transport of radionuclides is however dominated by diffusion. It has been noticed that the method used by REPCOM works well in these kind of cases. Several verifications against analytical formulas or other computer models have confirmed this.

⁹LAPACK = Linear Algebra PACKage provided, for example, by webpage (LAPACK, 2008). To understand the methods LAPACK uses, consult any proper book concerning numerical linear algebra, for example (Trefefen and Bau III, 1997).

5 DEVELOPING THE TRANSPORT MODEL

5.1 The Geometry

A schematic presentation of the modelled deposition hole is shown in Figure 5-1. The damage of the canister is assumed to be severe, and it is presented as a 35 cm high missing part of the canister at the top of it. The part corresponds the inner wall of the buffer block B1. In the model, ground water flows in a rock fracture spreading into the disturbed rock zone next to outer wall of the buffer block B1. The equivalent rate of this flow is denoted with Q_F . Respectively, the equivalent ground water flows from the backfill in the deposition hole and the tunnel section are Q_{DZ} and Q_{TDZ} , respectively. Some conservative assumptions are made, like the half a meter high missing buffer part under the canister. The geometry and some data of the deposition environment in numbers are presented in Table 5-1. The material properties of the real nuclides have been compiled in Appendix B. In all the computed cases, the bedrock environment is non-saline and all the values of the properties follow this assumption.

5.2 The Step by Step Developing Strategy

To make sure that the model under introduction will work properly, a careful step-by-step developing strategy is used. The idea is to include each new property in the model one by one, and compare the new results to the reference results obtained with another model. In this case, GoldSim is developed according to the strategy with REPCOM as a reference model. The REPCOM results were computed by Henrik Nordman, following the approach described in (Nordman and Vieno, 2003).

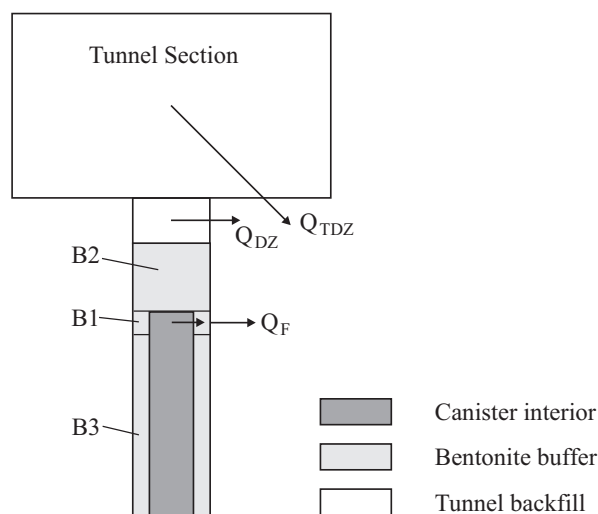


Figure 5-1. A side view of the modelled deposition hole (Nordman and Vieno, 2003).

Table 5-1. *The geometry and data of the model. (Nordman and Vieno, 2003)*

Canister interior	
fuel	2.14 tU
water volume	0.700 m ³
outer radius	0.53 m
fuel matrix degradation rate	10 ⁻⁶ 1/year
Buffer block B1	
height	0.35 m
inner radius	0.53 m
outer radius	0.88 m
equivalent water flow from the outer surface Q_F	0.2 l/year
Buffer block B2	
height	1.5 m
radius	0.88 m
Buffer block B3	
height	4.5 m
inner radius	0.53 m
outer radius	0.88 m
Backfill in the upper part of the deposition hole	
height	1.0 m
radius	0.88 m
equivalent water flow from the outer surface Q_{DZ}	2 l/year
Backfill in the tunnel section	
volume	100 m ³
equivalent water flow from the tunnel Q_{TDZ}	10 l/year

The first step is to test the GoldSim with verification nuclides that have properties of real nuclides in different bedrock environments. These nuclides do not decay, and thus they test, if the mass transport properties of the program work properly. These properties include diffusion, retardation, solubility, and general properties like geometry. The step corresponds to the comparison between REPCOM and Porflow done in (Nordman and Vieno, 2003). The grid that is used follows the precision of the grid used in REPCOM, and it is drawn in Figure 5-2(a).

The next step is to test the decay with a short decay chain. Also, the shared solubility is tested with two isotopes of the same element. After this stage all the basic properties have been tested, and it is time to proceed to use the dense grid drawn in Figure 5-2(b). Finally, several decay chains are tested simultaneously and the results compared with the separate REPCOM calculations.

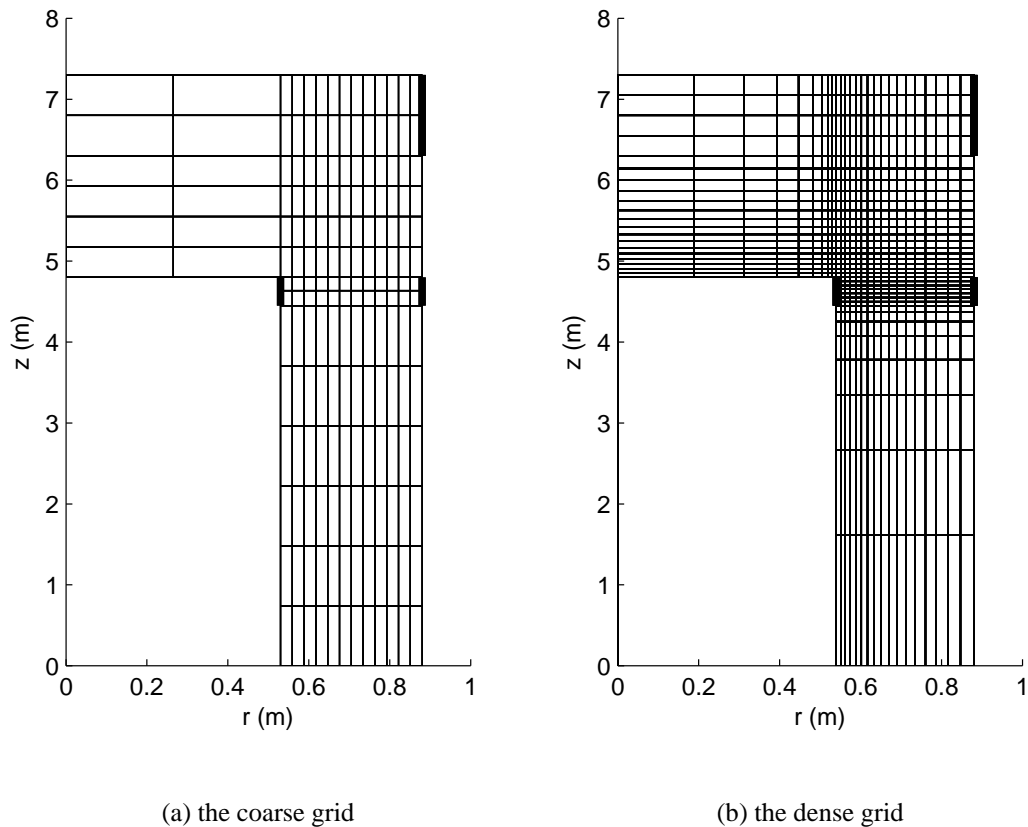


Figure 5-2. The grids in r - z -coordinates. The details of the grids can be seen in Appendix A. The bold lines on the outer edge present the flow boundary conditions caused by the equivalent flows Q_F and Q_{DZ} . The inner bold line denotes the length of the canister where the damage has occurred, i.e. there is no wall at that location. The uppermost cells with the upper wall at height 7.3 m neighbour the tunnel where the nuclides can diffuse. The tunnel is modelled as a separate volume, and the equivalent flow from the tunnel is Q_{TDZ} . All the other boundaries are impenetrable. The diffusive lengths in the canister and in the tunnel have been conservatively estimated to be negligible, i.e., zeros.

5.3 The Tests with the Stable Verification Nuclides

The stable verification nuclides are of different types: non-sorbing neutral N-S, sorbing neutral S, anion A, and cation C. The material properties for these nuclides are shown in Table 5-2. Two tests were performed for these nuclides. In the first one, an instant release of one mole of each nuclide without solubility limits was simulated. The release rate from the near-field, i.e. the rate the nuclides diffuse to the flows Q_F , Q_{DZ} , and Q_{TDZ} , is plotted in Figure 5-3(a). The second test probes the performance of GoldSim in presence of solubility-limited species. The solubility is limited to 1 mol/l for all nuclides, and the instant release is 10^9 moles to make sure that the solubility is limited inside the canister from the beginning. The results are presented in Figure 5-3(b). For the non-

Table 5-2. *The verification nuclides*

	N-S	S	A	C
Speciation	Neutral	Neutral	Anion	Cation
Buffer (non-saline)				
K_d (m ³ /kg)	0	0.3	0	0.018
ε	0.43	0.43	0.17	0.43
D_e (m ² /s)	$1.2 \cdot 10^{-10}$	$1.2 \cdot 10^{-10}$	$1 \cdot 10^{-11}$	$3 \cdot 10^{-10}$
Backfill (non-saline)				
K_d (m ³ /kg)	0	0.3	0	0.0061
ε	0.23	0.23	0.23	0.23
D_e (m ² /s)	$2 \cdot 10^{-10}$	$2 \cdot 10^{-10}$	$2 \cdot 10^{-10}$	$2 \cdot 10^{-10}$

solubility-limited case, the maximum rates and their times of occurrence can be found in Table 5-3. Also, the equilibrium rates in the limited case have been collected into the same table.

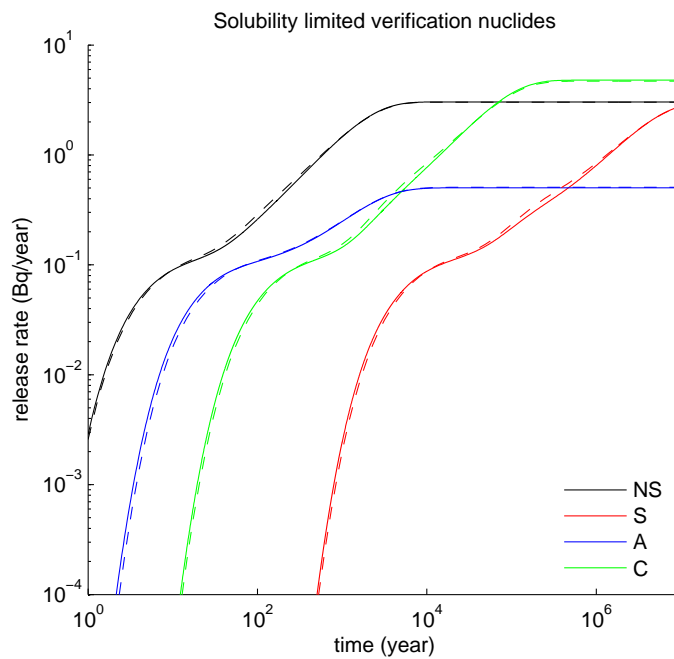
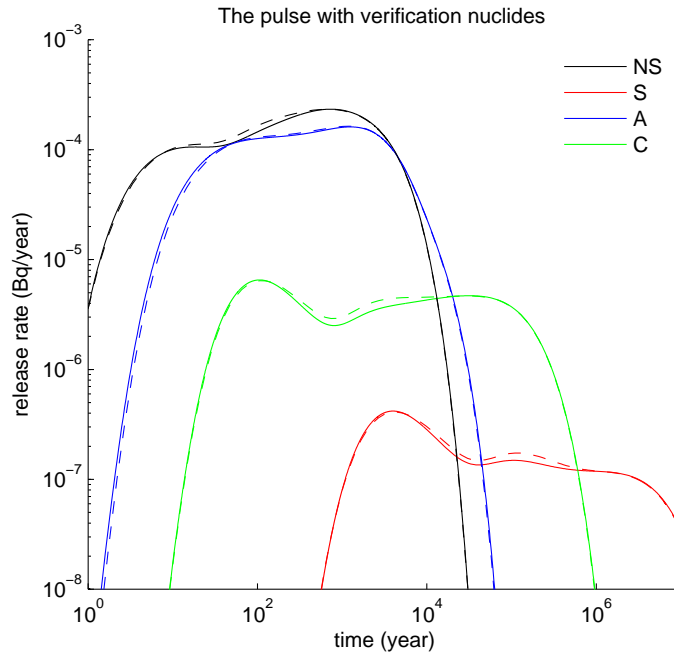


Figure 5-3. The result with the verification nuclides. The solid lines present the GoldSim results and the dashed line the REPCOM results. The upper figure is the 1 mole instant release case without solubility limit. The lower figure is the solubility limited case with the large instant release. Some minor differences can be observed especially at the mid-stages of the plots.

Table 5-3. The maximum rates and the maximum times for the case with no the solubility limits as well as the limits and the times when 90 per cent of the limits are reached for the limited case.

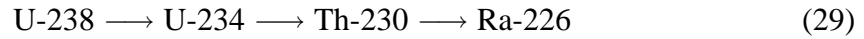
The non-solubility-limited case with one mole inside canister at $t=0$				
nuclide	time (year)		maximum (mol/year)	
	GoldSim	REPCOM	GoldSim	REPCOM
N-S	$7.29 \cdot 10^2$	$6.30 \cdot 10^2$	$2.34 \cdot 10^{-4}$	$2.33 \cdot 10^{-4}$
S	$3.99 \cdot 10^3$	$4.36 \cdot 10^3$	$4.19 \cdot 10^{-7}$	$4.10 \cdot 10^{-7}$
A	$1.24 \cdot 10^3$	$1.20 \cdot 10^3$	$1.61 \cdot 10^{-4}$	$1.64 \cdot 10^{-4}$
C	$1.06 \cdot 10^2$	$1.06 \cdot 10^2$	$6.53 \cdot 10^{-6}$	$6.43 \cdot 10^{-6}$

The solubility limited case				
nuclide	time when 90% is reached (year)		limit rate (mol/year)	
	GoldSim	REPCOM	GoldSim	REPCOM
N-S	$3.62 \cdot 10^3$	$3.71 \cdot 10^3$	3.04	3.03
S	$>1 \cdot 10^7$	$>1 \cdot 10^7$	2.78*	2.76*
A	$4.48 \cdot 10^3$	$5.13 \cdot 10^3$	0.503	0.508
C	$1.7 \cdot 10^5$	$1.69 \cdot 10^5$	4.79	4.69

*The value at the end of the computation that is at 10^7 years. The real maxima will be reached later and they will have the same values as the non-sorbing nuclide N-S.

5.4 A Simple Decay Chain with Shared Solubilities

The non-decaying calculations presented above are followed by the computation of the decay chain



and separate Np-237 simultaneously with the coarse grid.¹⁰ In addition, the uranium isotopes share the solubility. The inventories and the nuclide related data can be found in Appendices B and C. The obvious next step is to compute the same nuclides with the dense grid. All the results obtained with REPCOM as well as coarse and dense grid with GoldSim have been plotted in Figure 5-4.

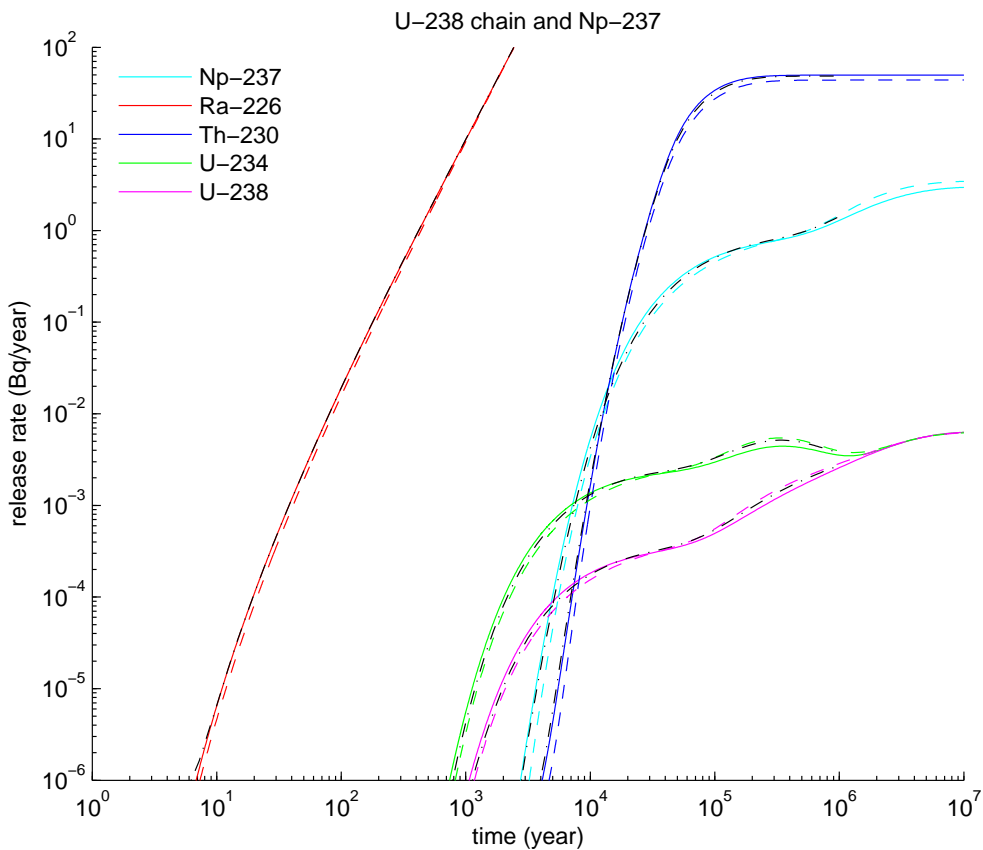
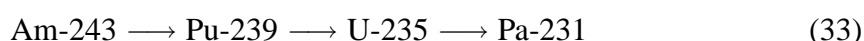
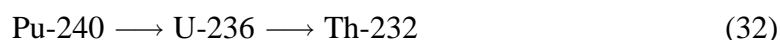
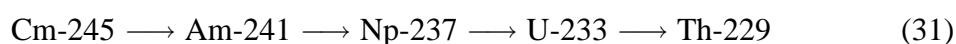
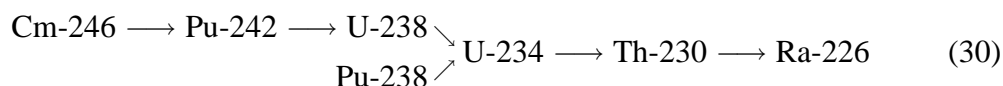


Figure 5-4. The release rate of the U-238 short chain (29) and Np-237. The solid coloured lines are the GoldSim dense grid results, and the dashed coloured lines are the GoldSim coarse grid results. The black dash-dot lines are the REPCOM results corresponding the nuclides of the GoldSim results nearby. The results agree with each other with only small differences.

¹⁰The nuclides with short half-lives have been neglected in this chain and also in the following chains .

5.5 Several Chains Computed Simultaneously

After the short chain and Np-237 computations, it is time to proceed to computing the chains



simultaneously with GoldSim and dense grid. The inventory and other data of the chains are shown in Appendices B and C. In the REPCOM modelling, the decay chains have been simplified in order to optimize the use of computing resources and to facilitate the treatment of elements appearing in several decay chains. The contribution formed by the relatively short-living parent nuclides (Pu 240; Cm-245, Pu-241, Am-241; Cm-246, Pu-242, Pu-238; Am-243, Pu-239) are added to the inventories of the daughters. To be precise, this means that the activity inventories of U-234, U-235, U-236, Np-237, and Am-241 at the sealing time of the repository are replaced by their respective maximum inventories appearing actually later. All plutonium isotopes were calculated simultaneously, which means that their solubility is treated correctly inside the canister. The other elements (Am, Cm, Th, U) appearing in two or more decay chains are handled separately using conservative simplifications.

Consequently, with REPCOM the following nuclide groups and decay chains are computed independently:

- Pu isotopes, U-236, Am-243 and Cm-246 as single nuclides
- chain Cm-245 \longrightarrow Am-241
- chain Np-237 \longrightarrow U-233 \longrightarrow Th-229
- chain U-238 \longrightarrow U-234 \longrightarrow Th-230 \longrightarrow Ra-226
- chain U-235 \longrightarrow Pa 231.

The results are presented in Figure 5-5. The nuclides that have clear maximums during the calculation time have been collected into Table 5-4 showing the maximums and the maximum times. Due the differences in the behaviour between GoldSim and REPCOM, the uranium isotopes have been left out from the table.

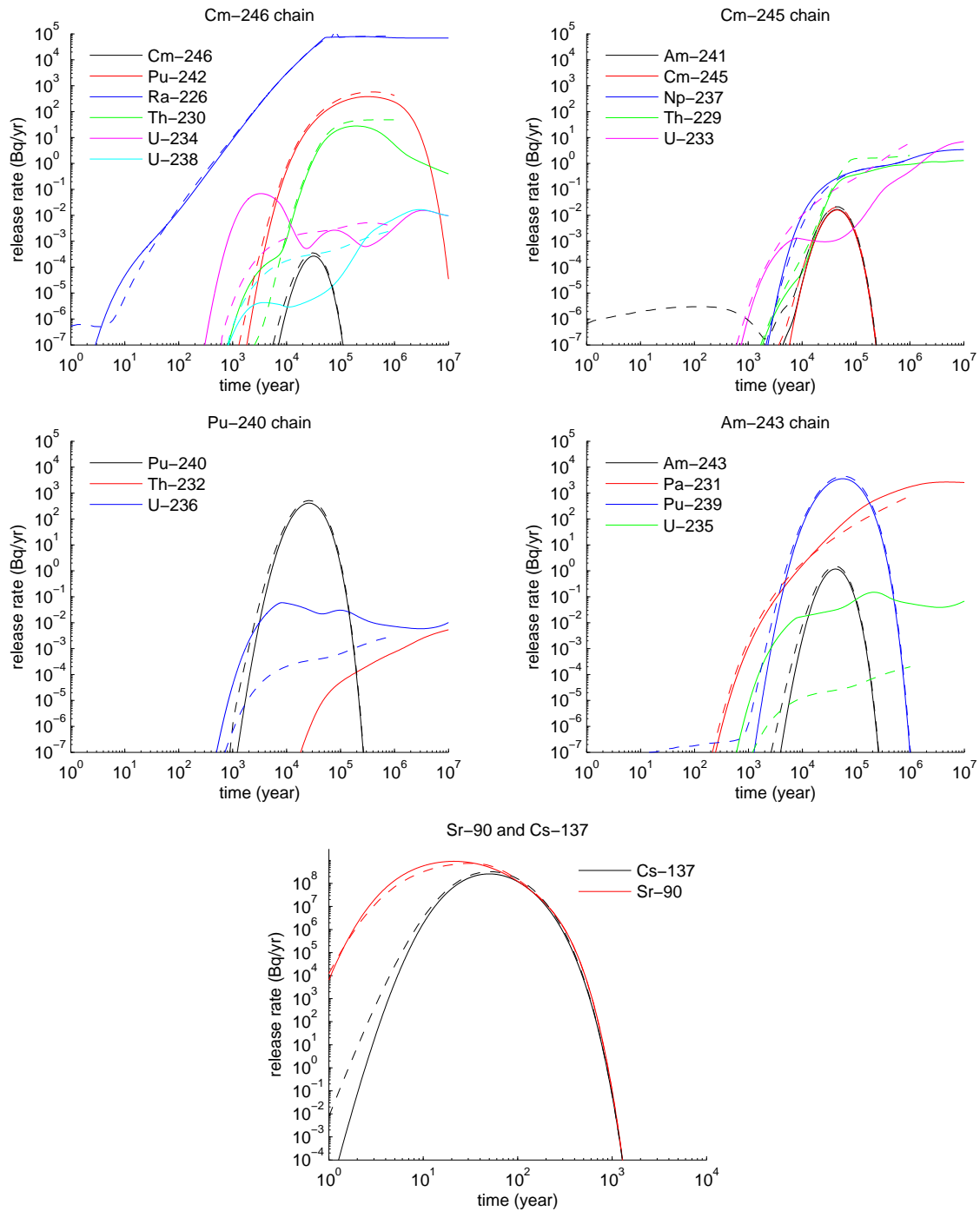


Figure 5-5. The release rates of the long chains (30)-(33) as well as Cs-137 and Sr-90. The GoldSim results are marked with solid lines and the REPCOM results with dashed lines. The purpose of these figures is rather to demonstrate the coupled effects of the nuclides of the same element than to be a precise comparison of the two models with different conservative estimates of the inventories.

As it can be observed from the plots, the results are somewhat different in particular at short times. However, the differences can be explained and are, in fact, expected. For example, the Pu-238 inventory in the GoldSim calculation causes the first 'spike' in the

Table 5-4. *The maximums of the multichain computation. Only the clear maximums are presented. The uraniums have been left out, because of the differences in the behaviour between the models used.*

nuclide	times (year)		maximums (Bq/year)	
	GoldSim	REPCOM	GoldSim	REPCOM
Am-241	$4.50 \cdot 10^4$	$4.17 \cdot 10^4$	$1.68 \cdot 10^{-2}$	$2.15 \cdot 10^{-2}$
Am-243	$4.10 \cdot 10^4$	$5.75 \cdot 10^4$	$1.12 \cdot 10^0$	$1.52 \cdot 10^0$
Cm-245	$4.50 \cdot 10^4$	$4.17 \cdot 10^4$	$1.60 \cdot 10^{-2}$	$2.04 \cdot 10^{-2}$
Cm-246	$3.20 \cdot 10^4$	$3.69 \cdot 10^4$	2.67^{-4}	$3.57 \cdot 10^{-4}$
Pu-238	$1.66 \cdot 10^3$	-	$7.0 \cdot 10^{-11}$	-
Pu-239	$5.60 \cdot 10^3$	$5.75 \cdot 10^4$	$3.59 \cdot 10^3$	$4.52 \cdot 10^3$
Pu-240	$2.60 \cdot 10^4$	$2.57 \cdot 10^4$	$4.08 \cdot 10^2$	$5.20 \cdot 10^2$
Pu-242	$3.20 \cdot 10^5$	$3.02 \cdot 10^5$	$3.80 \cdot 10^2$	$5.75 \cdot 10^2$
Ra-226	$2.30 \cdot 10^5$	$7.94 \cdot 10^4$	$7.88 \cdot 10^4$	$1.31 \cdot 10^5$
Th-230	$2.00 \cdot 10^5$	$3.49 \cdot 10^5$	$2.79 \cdot 10^1$	$4.80 \cdot 10^1$
Cs-137	$4.90 \cdot 10^1$	$5.53 \cdot 10^1$	$2.60 \cdot 10^8$	$3.26 \cdot 10^8$
Sr-90	$2.11 \cdot 10^1$	$3.38 \cdot 10^1$	$9.14 \cdot 10^8$	$7.47 \cdot 10^8$

U-234 curve and the 'bulge' in the Th-230 GoldSim graph. Plutonium is thousand times more soluble than uranium and the distribution coefficient of plutonium is 4 (m^3/kg) compared to the uranium's 0.5 (m^3/kg), making the release of the nuclides appear much earlier in the GoldSim results than in the REPCOM ones. Also, in GoldSim results, Th-230 release is decreasing later due to ingrowth of Th-232 which dominates the solubility limit. The ingrowth of Th-232 from U-236 was not included in REPCOM calculations. Similarly, the other nuclides that have been simplified in the REPCOM calculations are the cause of the differences in the release rates of their daughter nuclides.

The overall behaviour of the uraniums is also worth commenting. At first sight the somewhat strange results can be explained by the coupled effects in the simultaneous GoldSim calculation. It must also be remembered that REPCOM takes the solubility limits into account only in the disposal canister i.e. not in the bentonite nor backfill. That is significant for the nuclides that have parents producing a significant amount of the nuclide outside the canister, so that these nuclides will have to precipitate in the bentonite. This kind of nuclides are, for example, uraniums and thoriums. To be more exact, this behaviour is seen with U-233, for instance. The solubility of the nuclide is limited in the bentonite in the GoldSim calculation, whereas with REPCOM it is not.

If the differences in the uranium isotopes between GoldSim and REPCOM are disregarded, the results of the other nuclides with high solubilities and short half-lives coincide rather well. Finally and most importantly, the nuclides causing the activity releases,

especially Ra-226, behave similarly. Also, the tests with the partly instant released, very soluble, and almost non-sorbing Cs-137 and Sr-90 in Figure 5-5 show that the transient behaviours are not too different.

6 DISCUSSION

6.1 Comparing GoldSim and REPCOM

The old program REPCOM works, but the new program GoldSim has some new useful and crucial features for realistic computing. First of all, the already mentioned user interface of GoldSim is much easier, more user-friendly, more illustrative and clearer than the one of REPCOM that it is alone good enough reason to begin using GoldSim instead of REPCOM. In addition, GoldSim contains many features that have not yet been introduced and that can be included into the model with ease. On the contrary, all the extra features in REPCOM should be programmed from scratch. This is significant when further developing of the model is considered.

The main reason to begin to use GoldSim is the somewhat unrealistic treatment of solubility limits on REPCOM. The lack of the limits in the bentonite and in the backfill along with the uncoupling of the nuclides of the same element pose a threat to reliable results. In the case studied in this report, the effects of these deficiencies on the radionuclides significant to long-term safety (Ra-226 and the plutonium isotopes)¹¹ are minor. However, if the chemical environment of the disposal site change, coupling the nuclides of same elements and limiting the solubility outside the disposal may cause the results to differ substantially. That is why the GoldSim model or a model with the same kind of coupling and solubility features should be used from now on, despite the increasing of the computing times. In addition, the price of the extra computation time caused by the dense grid is worth paying to make sure the results are precise enough. On the other hand, the model may need some fine-tuning, if the number of the cases increase significantly or Monte Carlo simulations are needed.

6.2 Future Prospects

The natural way to continue with the model is to compute the release rates when the whole inventory in the separate parts of the fuel rods are taken into account. The whole inventory case can be extended to the other cases with different waste types, and further to the cases with different groundwater and bedrock environments, i.e. to the cases with different chemical properties. Also, the size of the damage to the canister can be varied, the equivalent flows in the bedrock changed, the time of failure can be assumed to follow some kind of probability distribution, the degradation rates of the parts where the nuclides are bound can be differed, and so on. The options are various, and the ones seen

¹¹The assesment of the nuclides causing the radiation risks is based on nuclide behaviour in the bedrock outside the disposal site. Calculation on this subject can be viewed in (Vieno and Nordman, 1999)

necessary should all be covered. One possibility to get a picture of the overall situation is to utilize the Monte Carlo simulation properties of GoldSim, if this kind of approach seems reasonable.

Another way to develop the model is to step outside the near-field to the bedrock and model the properties of the rock matrix. GoldSim has features that allow this. For example, it has tools to model the sorption of the radionuclides into the fracture zones of the rock matrix during the transport through the bedrock. However, this kind of development is near the groundwater flow modelling and should be done in co-operation with the researchers of the field. They may have better tools to build the model, or at least they can comment on the GoldSim approach.

An alternate way to advance the model is to describe the physical phenomena in a more accurate manner. For example, chemical reactions between the nuclides and other particles could be included. Also, an accurate description of sorption and other surface phenomena could be calculated more precisely than now. However, these additions would increase the computing time substantially and may be worth while just for some test cases.

Cylindrical geometry without azimuthal symmetry is easy to use and fast to compute with. However, a real three dimensional model should be made to validate the results obtained with the cylindrical model. The cylindrical coordinates make it difficult to handle some of the failure cases. Modelling a defect that is located in a geometrically asymmetric position about the z -axis is not possible in a realistic way. In addition, the effects of the groundwater flowing only in one direction in a narrow fracture should be studied. The problem with a 3D model is that it requires significant computational power. That is why it should be applied only to few simple cases, at least in the beginning. A program with which the 3D model could be done is COMSOL Multiphysics. An advantage of the COMSOL is that it is a finite element software, and thus the results would be achieved with different method thus adding reliability.

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APPENDICES

A GRID AND TIMESTEP DETAILS

Table A-1. Timestep Details

Time range (year)	# steps	step length (year)
0-10 ³	10 ⁴	0.1
10 ³ -10 ⁴	9·10 ³	1
10 ⁴ -10 ⁵	9·10 ³	10
10 ⁵ -10 ⁶	9·10 ³	100
10 ⁶ -10 ⁷	9·10 ³	1000

Table A-2. The GoldSim precision settings. The settings that were used are low for the verification nuclides and high for the other cases. The change in isotope ratio means the change that causes GoldSim to relinearize the unidirectional diffusive flow. (GoldSim, 2007)

Precision Setting	Low	Medium	High
Change in isotope ratio	0.50	0.30	0.10
Solubility limit overshoot	0.10	0.05	0.025
Fractional timesteps	1	4	10
Solver tolerance	1·10 ⁻⁶	1·10 ⁻⁷	3·10 ⁻⁹
Solver mass tolerance (g)	1·10 ⁻¹⁷	1·10 ⁻¹⁹	1·10 ⁻²²
Shortest timestep fraction	1·10 ⁻⁶	1·10 ⁻⁷	1·10 ⁻⁸

Table A-3. Grid points. The REPCOM grid above $z=6.3$ m is divided radially only into two parts: 0-53cm and 53-88 cm.

	Coarse grid		Dense grid		REPCOM grid		
	r (cm)	z (m)	r (cm)	z (m)	r (cm)	z (m)	
0	0	0	0	0	0	0	0
1	26.5	0.742	18.80482	1.618726	53	1.11	1
2	53	1.483	31.17086	2.666814	56.18	2.23	2
3	55.91667	2.225	39.30277	3.345427	59.36	3.34	3
4	58.83333	2.966	44.65031	3.784814	62.55	4.45	4
5	61.75	3.708	48.16684	4.069307	65.73	4.80	5
6	64.66667	4.45	50.47932	4.25351	68.91	5.18	6
7	67.58333	4.635	52	4.372777	72.10	5.55	7
8	70.5	4.8	53	4.45	75.27	5.93	8
9	73.41667	5.175	54	4.5	78.45	6.30	9
10	76.33333	5.55	55.07301	4.55	81.64		10
11	79.25	5.925	56.22438	4.6	84.82		11
12	82.16667	6.3	57.4598	4.65	88		12
13	85.08333	6.8	58.78543	4.7			13
14	88	7.3	60.20786	4.75			14
15			61.73414	4.8			15
16			63.37186	4.85			16
17			65.12916	4.903936			17
18			67.01477	4.962118			18
19			69.03805	5.024881			19
20			71.20907	5.092584			20
21			73.5386	5.165617			21
22			76.03822	5.244399			22
23			78.72035	5.329383			23
24			81.59832	5.421057			24
25			84.68642	5.519949			25
26			88	5.626625			26
27				5.741699			27
28				5.865832			28
29				5.999736			29
30				6.144183			30
31				6.3			31
32				6.55			32
33				6.8			33
34				7.05			34
35				7.3			35

B RADIONUCLIDE AND MATERIAL PROPERTIES

Table B-1. The properties of the radionuclides. The distribution coefficients K_d 's are between the solids (bentonite and backfill) and water. (SKB, 2006a)

Nuclide	Half-life (year)	K_d (m ³ /kg)		
		Bentonite	Backfill	solubility (mol/l)
Am-241	$4.3 \cdot 10^2$	10	3.2	$4.0 \cdot 10^{-7}$
Am-243	$7.4 \cdot 10^3$	10	3.2	$4.0 \cdot 10^{-7}$
Cm-245	$8.50 \cdot 10^3$	10	3.1	$4.0 \cdot 10^{-7}$
Cm-246	$4.70 \cdot 10^3$	10	3.1	$4.0 \cdot 10^{-7}$
Cs-137	$3.0 \cdot 10^1$	0.018	0.0061	∞
Np-237	$2.10 \cdot 10^6$	4	1.2	$1.1 \cdot 10^{-9}$
Pa-231	$3.2 \cdot 10^4$	0.2	0.095	$3.0 \cdot 10^{-7}$
Pu-238	$8.80 \cdot 10^1$	4	1.3	$1.1 \cdot 10^{-6}$
Pu-239	$2.4 \cdot 10^4$	4	1.3	$1.1 \cdot 10^{-6}$
Pu-240	$6.5 \cdot 10^3$	4	1.3	$1.1 \cdot 10^{-6}$
Pu-242	$3.8 \cdot 10^5$	4	1.3	$1.1 \cdot 10^{-6}$
Ra-226	$1.60 \cdot 10^3$	0.001	0.007	$2.2 \cdot 10^{-8}$
Sr-90	$2.9 \cdot 10^1$	0.0009	0.00028	$9.1 \cdot 10^{-5}$
Th-229	$7.30 \cdot 10^3$	6	1.9	$6.3 \cdot 10^{-9}$
Th-230	$7.70 \cdot 10^4$	6	1.9	$6.3 \cdot 10^{-9}$
Th-232	$1.4 \cdot 10^{10}$	6	1.9	$6.3 \cdot 10^{-9}$
U-233	$1.60 \cdot 10^5$	0.5	0.15	$9.5 \cdot 10^{-10}$
U-234	$2.40 \cdot 10^5$	0.5	0.15	$9.5 \cdot 10^{-10}$
U-235	$7.0 \cdot 10^7$	0.5	0.15	$9.5 \cdot 10^{-10}$
U-236	$2.3 \cdot 10^7$	0.5	0.15	$9.5 \cdot 10^{-10}$
U-238	$4.50 \cdot 10^9$	0.5	0.15	$9.5 \cdot 10^{-10}$

Table B-2. Buffer and backfill properties. All the nuclides, except cesium, are neutral in the assumed dilute water environment. (SKB, 2006a)

Speciation	neutral	anion	Cs
Buffer			
ε	0.43	0.17	0.43
D_e (m ² /s)	$1.2 \cdot 10^{-10}$	$1 \cdot 10^{-11}$	$3 \cdot 10^{-10}$
Grain density (kg/m ³)	2700	2700	2700
Backfill			
ε	0.23	0.092	0.23
D_e (m ² /s)	$5 \cdot 10^{-11}$	$4.2 \cdot 10^{-12}$	$1.3 \cdot 10^{-10}$
Grain density (kg/m ³)	2700	2700	2700

C THE CONTENTS OF THE WASTE

Table C-1. The radionuclide inventory of a single canister containing BWR fuel from the OL1 and OL2 reactors after 30 years of cooling. The assumed burnup is 40 MWd/kgU and enrichment 4.2% (Anttila, 2005)

nuclide	activity at 30 year (Gbj/tU)	Partitioning	
		fuel matrix	instant release
Am-241	$1.41 \cdot 10^5$	1	0
Am-243	$7.62 \cdot 10^2$	1	0
Cm-245	6.16	1	0
Cm-246	1.19	1	0
Cs-137	$2.36 \cdot 10^6$	0.95	0.05
Np-237	$1.30 \cdot 10^1$	1	0
Pa-231	$1.40 \cdot 10^{-3}$	1	0
Pu-238	$8.75 \cdot 10^4$	1	0
Pu-239	$1.05 \cdot 10^4$	1	0
Pu-240	$1.99 \cdot 10^4$	1	0
Pu-242	$7.62 \cdot 10^1$	1	0
Ra-226	$3.50 \cdot 10^{-4}$	1	0
Sr-90	$1.70 \cdot 10^6$	0.99	0.01
Th-229	$1.00 \cdot 10^6$	1	0
Th-230	$1.60 \cdot 10^{-2}$	1	0
Th-232	0	1	0
U-233	$2.41 \cdot 10^{-3}$	1	0
U-234	$5.59 \cdot 10^1$	1	0
U-235	$7.48 \cdot 10^{-1}$	1	0
U-236	$1.30 \cdot 10^1$	1	0
U-238	$1.16 \cdot 10^1$	1	0