

Comparison of different SFL design alternatives

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Summary

Four different design options for a repository for long-lived nuclear waste from the future dismantling of the nuclear power plants have been compared. The time scales considered range up to 100,000 years. The repository is to be located at about 500 m depth in granitic rock. The vault can be a tunnel about 200 m long and on the order of 15×15 meters, in which the waste is surrounded by either a hydraulic cage, a concrete buffer or a bentonite buffer about 2 m thick. A fourth option is to make a silo, called Supersilo, about as high as wide, surrounded by both concrete and bentonite.

In order to compare potential release rates of radionuclides from the waste to the seeping water in the rock a number of simple models have been devised. Some of these models allow the *water flow rates through vaults* to be assessed under various conditions and configurations. Other models are used to calculate the uptake by molecular diffusion to the water in the rock *that seeps past* the vaults. Moreover other models are used to calculate the rate of transport of nuclides by *diffusion and flow through* the buffer and waste. The decay of the nuclides during their passage from the waste to the flowing water through and past the vaults is accounted for. Many nuclides of interest decay considerably in the buffer. The mathematical form of the models is made so simple that essentially hand calculations can be used to explore the strength of different barriers and design options. The simple models are validated against more complex coupled models accounting for simultaneously competing processes. The more complex models are solved by numerical methods. The toolbox of simple models is used to calculate the strength of the barriers in the different design options under various conditions. Examples of activity releases of three nuclides with different sorption characteristics and half-lives are presented.

It is found that a hydraulic cage is not a good option as it promotes the release of nuclides instead of hindering the release. The concrete barrier in the vault is the strongest, both because of its low hydraulic conductivity and its good sorption properties, provided it does not fracture or degrade seriously. Bentonite allows more water to flow through the buffer and waste and to contaminate the water in the rock because it has higher hydraulic conductivity and lower retardation of the nuclides. However, it may be less prone to developing fractures than concrete. The silo option is somewhat better whichever buffer is used because it allows less water to flow through the silo. The silo with both concrete and bentonite, with bentonite outermost, has the strongest retention properties because even if concrete should be considerably fractured the bentonite hinders a large increase in flow through the buffers. This arrangement also retains the good retardation in the concrete. However, concrete and bentonite in contact mutually degrade each other, given time. Ordinary Portland cement based concrete in contact with bentonite could mutually degrade up to a metre over 100,000 years. Low pH cement would limit the degradation by chemical interaction to some ten(s) of centimetres.

Sammanfattning

Fyra olika konstruktionsalternativ för ett slutförvar för långlivat radioaktivt avfall från framtida nedmontering av kärnkraftverk har jämförts. Utsläppstider av radioaktivitet upp till 100 000 tals år har studerats. Förvaret ska förläggas på cirka 500 m djup i granitiskt berg. Valvet kan vara en ca 200 m lång tunnel med ett tvärsnitt av cirka 15×15 meter, där avfallet omges av antingen en hydraulisk bur, en betongbuffert eller en bentonitbuffert, cirka 2 m tjock. Ett fjärde alternativ är att göra en silo, kallad Supersilo, ungefär lika hög som bred, omgiven av både betong och bentonit.

För att kunna jämföra eventuella utsläppshastigheter av radionuklider från avfallet till sipprande vattnet i berget har ett antal enkla modeller tagits fram. Några av modellerna används för att beräkna vattenflödet genom valven under olika förhållanden och utföranden. Andra modeller används för att beräkna utsläppet via molekylär diffusion till vattnet som sipprar i berget. Andra modeller återigen används för att beräkna transporthastigheten av nuklider genom diffusion och flöde genom buffert och avfall. Nuklidavklingningen under passagen från avfallet till det strömmande vattnet i berget beaktas i modellerna. Många nuklider av intresse avklingar väsentligt i bufferten. Modellerna har gjorts så enkla att i huvudsak handberäkningar kan användas för att undersöka styrkan i olika barriärer och av alternativa utformningar av förvaren. De enkla modellerna har validerats mot mer komplexa kopplade modeller där många samtidigt konkurrerande processer beaktas. De mer komplexa modellerna kräver användning av mer omfattande numeriska metoder. De enkla modellerna används för att beräkna barriärernas effekt i olika designalternativ och under olika förhållanden. Exempel på utsläpp av tre nuklider med olika sorptionsegenskaper och halveringstider presenteras.

Det visar sig att en hydraulisk bur inte är ett bra alternativ eftersom det främjar frisättning av nuklider i stället för att motverka utsläpp. Betongbarriären är den starkaste, både på grund av den låga hydrauliska konduktivitet som av de goda sorptionsegenskaperna, förutsatt att betongen inte spricker eller bryts ned kraftigt. Bentonit tillåter mer vatten att passera genom bufferten och avfallet och vattnet i berget omkring förvaret blir mer förorenat eftersom bentonit har högre hydraulisk konduktivitet och ger lägre fördröjning av radionuklider. Emellertid kan det vara mindre benäget att utveckla sprickor än betong. Siloalternativet är något bättre än tunnelalternativet beroende på att det tillåter mindre vatten att strömma genom silon. Silon med både betong och bentonit, med bentonit ytterst, har de starkaste retentionsegenskaperna eftersom även om betongen skulle utveckla sprickor så hindrar bentoniten en stor ökning i flödet genom buffertarna. Detta arrangemang bibehåller också den goda fördröjningen av nukliderna i betongen. Emellertid försämrar betong och bentonit i kontakt ömsesidigt varandras egenskaper. Ordinarie Portlandcement-baserad betong i kontakt med bentonit skulle ömsesidigt försämrade varandras egenskaper upp till en meter över 100 000 år. Låg-pH cement skulle begränsa förändringarna genom kemisk interaktion till någon/några tiotal centimeter.

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1 Introduction and background

A repository for long-lived low and intermediate level waste, SFL, is planned to be constructed in the future. The long-lived low and intermediate level waste consists of:

- Historical waste from AB Svafo and Studsvik Nuclear, which is mostly in drums filled waste and grout.
- Waste from the Swedish nuclear power plants in the form of:
 - the BFA tanks, which are painted steel tanks 3,300 mm long, 1,300 mm wide and 2,300 mm high with about 12,000 kg waste,
 - cubical concrete moulds with sides 1.2 m each weighing at most 5,000 kg.

The waste is sufficiently active and long-lived to locate the waste repository at 300–500 m depth in granitic rock. However, the waste is not so active that it needs to be encased in extremely long-lived canisters, as does the high level waste. It is deemed to be sufficient to ensure that the water flowrate through the waste or alternatively the release rate to groundwater that flows around the repository surrounded by some diffusion barrier is sufficiently low to ensure safety.

Two main different concepts, hydraulic cage and barriers, are contemplated. In the first, called a hydraulic cage, the waste is emplaced in vaults and surrounded all around by a very permeable medium such as crushed rock. As the waste is much less permeable than the crushed rock, groundwater seeping in the rock will flow into the highly permeable region around the waste and only a small fraction will flow *through* the waste to carry nuclides with it. Nuclides will additionally be released to the water in the cage by molecular diffusion. The diffusion properties of the wastes and the crushed rock must also be considered. The hydraulic properties of the wastes and their development over time can become important and must be assessed. Also possible degradation and clogging of the crushed rock will influence the release. The design of the waste form and any enclosures can also affect the hydraulic and diffusive properties of the materials.

In the second main alternative the opposite conditions are strived for, namely to minimize the flow through the material surrounding the waste. The waste is surrounded by buffer material, e.g. compacted bentonite clay that has a much lower hydraulic conductivity than the surrounding rock. The idea is that water flowing in the rock is diverted around the buffer and only a fraction flows through it and through the waste. This water will carry dissolved nuclides. In addition the nuclides will diffuse through the buffer to be carried away by the water seeping in the fracture network in the rock. The mass transfer rate to the water in rock fractures must be considered as well as the diffusion rate through the buffer and the retardation by sorption. In contrast to the hydraulic cage case the clay buffer, if thick, can delay the diffusing nuclides during their passage through the buffer. Strongly sorbing nuclides with not very long half-lives may decay to insignificance before reaching the mobile water in the rock fractures. However, the buffer must remain in place and maintain its low hydraulic conductivity, low diffusion and high sorption properties over the time of interest, i.e. up to hundreds of thousands of years. A clay buffer can be degraded by reactions with the large amounts of very high pH concrete in the waste. Clay must also not erode away by low ionic strength water seeping or even flowing in fracture zones that intersect the vaults with the waste.

There are a number other factors that must be considered. Some of the nuclides are embedded in metal parts. They will be released at the rate at which the metals corrode. Gas that evolves by metal corrosion or possibly by the degradation of organic matter must be allowed to escape. The gas may carry volatile components with radioactive nuclides. The concrete waste containers and concrete supporting walls, if used, may at first be strong barriers to flow but may degrade by leaching and fracturing and eventually become more permeable.

Other factors that must be considered include the direction of water flow in the rock compared with the vaults. It may be perpendicular to the vault, along the vault or at any angle to the vault. The flowrate and direction may also change over time.

The following reports have given valuable background information: Skagius et al. (1999), Pettersson et al. (1999), Karlsson et al. (1999) and Holmén (1997).

2 Aims and scope of the SFL concept study

The overall aim is:

“to identify, describe and evaluate concept candidates for future repositories for long-lived low and intermediate level waste, SFL.”

For operational reasons we interpret this to mean that:

The main aim is to study and compare different design alternatives for SFL repositories from the point of view of long-term radioactive nuclide release to the seeping water in the rock. The results will be used to help to select a design that is sufficiently safe for the long-lived nuclear waste that will be deposited. To achieve this, simple models will be devised and used to compare different design alternatives. It is not the aim of the work to devise a full-fledged model that can be used to perform detailed release calculations for the large number of different nuclides. However, the simple models should be useful in subsequent studies to decide and select which mechanisms and processes should be incorporated in the more detailed models.

3 Approach to the task

As mentioned in the introduction there are many issues that must be considered in order to determine how the rate of release of nuclides to the seeping water in the rock will vary over time. As the main aim of this report is to supply sufficient and well-founded information to compare design options, we try to identify the main processes and mechanism. In this first step the two main options; the *hydraulic cage* and the *diffusion barrier* are modelled. Some simplifying assumptions are made on the hydraulic and diffusion properties of the rock, cage, buffer and waste, the properties of which are given a few discrete values in a base case. Hydraulic data of the site are largely based on conditions at Laxemar. The fractured rock is modelled as a porous medium instead of a more realistic fracture network. However, the impact of the presence of fractures and fracture zones on the mass transfer between seeping water and bentonite and concrete is accounted for.

Due to the long times that need to be considered the potential changes of the physical and chemical properties of the waste, bentonite and concrete need to be considered. Especially the potential increase in the hydraulic conductivity may lead to more rapid release of nuclides at later times. A new model that describes the mutual degradation of bentonite and concrete is used for this purpose.

Subsequently the simplifying methods are revisited in order to assess whether other assumptions could seriously influence the results. We have based the first simplifying assumptions on simple deliberations and on previous experience of what may be reasonably realistic and on the conservative side.

The report is structured in the following way. Chapter 4 summarises geometric, hydraulic and transport data for a base case. Section 5.2 outlines the conceptual models used to describe the different cases. In Section 5.2.3 the concept of and the basic equations for the equivalent flowrate Q_{eq} model are presented. This describes how nuclides are transferred by diffusion to the water *passing* the buffer and is central to the cases when there is little flow through the repository. Section 5.2.4 analyses cases when the buffer and waste have large hydraulic conductivity compared with the surrounding rock and allow much flow through the waste. This is useful for the study of cases when both the waste and buffer have a higher hydraulic conductivity than the rock. It allows us to consider “what-if” cases such as when for example concrete and waste have been severely fractured or degraded. Section 5.3 specifically addresses the “hydraulic cage” by using the Q_{eq} concept in combination with the model for flow through a high conductivity region surrounding a low conductivity waste. Section 5.4 uses the Q_{eq} concept to explore the impact of a prominent fracture and of a fracture zone that intersect the vault with *low conductivity waste as well as low conductivity barrier*. In Section 5.5 the impact of a prominent fracture or fracture zone on a *high conductivity waste as well as a high conductivity barrier* is explored. Section 5.6 summarises and discusses the cases presented in Chapter 5. The cases in this chapter describe the *carrying capacity of nuclides by the water* flowing in and around a repository vault.

In Chapter 6 the release from the waste and the nuclide transport through the bentonite and concrete barriers, which causes the release to the mobile water is discussed. Section 6.1 describes and quantifies how nuclides released from the waste form *diffuse* through a barrier subject to decay and retardation by sorption. Some simple but powerful analytical solutions are used to explore which nuclides can be expected to decay to insignificance during their migration through the buffer. In Section 6.2 these simple formulas are used to screen which nuclides need not be treated in detailed calculations. In Section 6.3 the instationary diffusion with sorption and decay in the buffer is combined with the Q_{eq} model for cases when there is *negligible flow through* the buffer and waste. This is a refinement of the previous case where the transport processes were treated as if they were independent. The diffusion resistance in waste, buffer and in the seeping water in the rock is accounted for in this case. The model is solved numerically and is somewhat more time consuming than the analytical solutions in previous sections. This approach gives less conservative results because it accounts for the interdependence of the different processes. In Section 6.4 sample calculations are presented for ^{137}Cs , ^{59}Ni and ^{226}Ra , three nuclides that potentially could be important in a safety analysis. In Section 6.5 a simple approach based on a combination of the Q_{eq} formulas and the diffusion through the barrier is tested against the model in 6.3. It is more conservative but is very simple and can possibly be used as an additional screening tool when one does not want to use numerical simulations.

In Chapter 7 a case is analysed where a low conductivity buffer embeds the waste with a much higher conductivity. This is conceivable e.g. for a clay buffer surrounding a fractured or degraded concrete waste. Section 7.2 describes such a case. It is found that considerable flowrates develop through the waste although it is embedded in the low conductivity buffer. This is especially pronounced when the hydraulic gradient is parallel to the vault. A model is presented which has a simple analytical solution. In Section 7.2 the flowrate through the waste and how the nuclides carried by the water are retarded and decay during the passage of the water out through the buffer is presented. Also here analytical solutions are used. In Section 7.4 some sample calculations are presented.

Chapter 8 discusses the mutual degradation of bentonite and concrete and presents some sample calculations using the model presented in Neretnieks (2013, manuscript to be published).

Chapter 9 makes comparison of and discusses the pros and cons of the different design options.

Chapter 10 discusses all the previous models and findings.

4 Basic data for the study under idealized conditions

4.1 Repository geometry

Several concepts with different barriers have been proposed. The different concepts are called:

- The gravel repository.
- The concrete repository.
- The clay repository.
- The Supersilo.

The *gravel repository* is essentially a tunnel in which the containers with waste are surrounded on all sides by gravel that acts as a hydraulic cage. Besides the waste containers themselves this is the only protective barrier aimed to divert the seeping groundwater around the waste to minimise flow through the waste. Nuclides will escape partly by flow through the waste and partly by diffusion from the waste to the water flowing in the gravel. Escape by diffusion will dominate in this case because the hydraulic cage will minimise flow through the waste.

In the *concrete repository* the waste containers are placed in a long, wide and high reinforced concrete construction, which is covered with a reinforced concrete roof. The remaining open space in the vault with the construction is poured full with concrete. As long as the concrete has not developed many fractures or been chemically degraded by leaching of soluble constituents to the seeping groundwater the concrete acts as a diffusion barrier. The more fractured the concrete walls become the more water can seep through and the more water will also flow through the waste inside. Should eventually concrete and waste inside it become more permeable than the surrounding rock all the water seeping through the waste may pick up nuclides. However, the degraded concrete may still retard some of the nuclides by sorption. pH will be maintained above 12 for a very long time if Portland cement is used.

The *clay repository* is similar to the concrete repository except that the space surrounding the construction is filled with (compacted) bentonite clay. As long as the bentonite is less permeable than the surrounding rock nuclide escape will be dominated by diffusion through it. Suitably chosen clays can retard many sorbing nuclides. pH will initially be between neutral and mildly basic. Some clay nearest the waste may degrade by reactions with hydroxyl ions diffusing out from the concrete in the waste. An additional concern will be erosion of bentonite by low ionic strength seeping water, should such water intrude during e.g. an ice age.

The *Supersilo* is not a tunnel but a vertical cylindrical reinforced silo with the waste, surrounded by first bentonite blocks, which in turn are encased in a second concrete silo surrounded by gravel on all sides. As long as the concrete walls have not degraded and the swelling bentonite clay also has not degraded and remains in place the release of nuclides to the seeping water in the rock is practically only by molecular diffusion through the barriers. Sorption retards the migration of most nuclides of interest in the clay as well as in the concrete. A variant of the Supersilo where the walls of silo are very thick and the bentonite is outside the silo wall is a further option. This was not proposed originally.

However, concrete and bentonite are chemically not compatible. Ordinary concrete releases alkali at high pH, first around 14 and later around 12.5. The hydroxyl ions and accompanying cations diffuse into the bentonite, which is a sink for them and the bentonite is degraded, diminishing its swelling, hydraulic and sorption properties. The concrete also degrades when much alkali has been lost and loses its strength and can conceivably become more like sand. Thick buffers will take longer time to degrade than thin.

In the first three cases the repository is made up of one or more vaults located at about 500 m depth in fractured granitic rock. We use the following simplified dimensions in this study, see Table 4-1. For the present simple modelling we need not use exact geometric data for the vaults and therefore simplify the repository dimensions. Having the same geometry makes it easier to see how the choice of barriers influences the result. Furthermore we can directly use some of the results presented in Holmén (1997) on how flowrates in the vaults and waste are influenced by hydraulic conductivity contrasts and flow angles.

Table 4-1. Dimensions of tunnels and waste.

Tunnel	Length 200 m Height 18 m Width 14 m This is equivalent of a cylinder radius of 8.96 m Volume 50,400 m ³
Waste	Length somewhat less than 200 m (196 m in example) Height 10 m Width 10 m This is equivalent of a cylinder radius of 5.64 m Radius when surrounded by 2 m concrete or clay buffer 7.64 m Volume of waste 20,000 m ³
Supersilo	Containing the same amount of waste as the tunnel Height 33.4 m Diameter 33.4 m Bentonite buffer is 2 m thick and Silo wall is also 2 m thick on all sides

We do not define the properties of the material between the outside of the buffer $r = 7.64$ m and the tunnel radius $r = 8.96$ m. In the release modelling cases it is assumed to have the same conductivity as the rock. The buffer volume in vault is 17,100 m³ and in the Supersilo the volumes of silo wall and the buffer together is 7,670 m³.

For later use we also define an ellipsoid that has the same volume as the tunnel. The ellipsoid is given the radii $r_b = r_c = 8.96$ m. Then the radius of the long axis $r_a = 150$ m which gives a 50% longer ellipsoid than the tunnel. The *largest* cross section seen in the direction along the main axis x is $A_{midx} = 252$ m². The area of the ellipse seen in the direction of the short axis y or z is $A_{midyz} = 4,220$ m².

4.2 Hydraulic (Kh , i), porosity (ϵ) and diffusion (D_p) data

The data are taken from Pettersson et al. (1999), SKB (2001), Höglund (2001), Skagius et al. (1999), and Ericsson et al. (2006). Sometimes the data differ between the reports. We have selected data we find reasonable based on previous experience. They are all within the range of published data. Waste and degraded concrete properties have been arbitrarily chosen for illustration purposes only.

The three nuclides Cl-36, Ni-59 and Ra-226 have different properties. Cl-36 has a long half-life and is not retarded by sorption in any of the media. Ni-59 has a fairly long half-life and moderate sorption coefficient in concrete as well as in bentonite. Ra-226 has a short half-life on geological time scales. It sorbs moderately well in concrete but less in bentonite. Data on other nuclide inventories, half-lives and retardation factors are compiled on Table 6-1.

Table 4-2 Data for the base case.

Data for sorption and retardation					
Mineral density	kg/m³				
Concrete	2,600				
Bentonite	2,600				
Porosity					
Construction concrete	0.15				
Bentonite	0.4				
Waste concrete	0.2				
Sorption coefficients m³/kg	Cl-36	Ni-59	Ra-226		
Concrete	0	0.04	0.05		
Bentonite	0	0.02	0.005		
Retardation factors					
Concrete	1	590	738		
Bentonite	1	79	21		
Halfives years					
	Cl-36	Ni-59	Ra-226		
	300,000	75,000	1,600		
Data for flow and diffusion					
	Hydraulic conductivity Kh m/s	Effective diffusion coeff De m²/s	Porosity ε	Pore diffusion coeff De/ε Dp m²/s	Buffer thickness m
Water	1.00E-09				
Rock	3.00E-09	1.00E-13	1.00E-04	1.00E-09	
Gravel	3.00E-06	5.00E-10	0.3	1.67E-09	2
Bentonite	1.00E-10	1.00E-10	0.4	2.50E-10	2
Construction concrete	5.00E-12	1.00E-11	0.15	6.67E-11	2
Degraded concrete	1.00E-10	1.00E-10	0.2	5.00E-10	2
Waste	1.00E-06	1.00E-10	0.2	5.00E-10	
Hydraulic gradient	3.00E-03				

5 Conceptual model for flow and solute transport under idealized conditions

5.1 Release from waste form

The nuclides in the waste are bound in metals, concrete, and ion exchange resins, most of them in concrete containers (1.2 m cube), which contain a cement waste mixture. Some nuclides exist as neutral or anionic species and can dissolve fully in the pore water in waste after water saturation. Some exist as cationic species. These sorb on or bind to ion exchange sites in cement and in bentonite. Some nuclides can form low solubility compounds. Some are bound in metal parts and will be released as the metals corrode. Over time, combinations of the dissolution and release mechanisms may change due to changes in the pore water chemistry. The quantification of these complex processes is outside the scope of this report. For simplification in the examples we use only constant release rate from the waste form or constant sorption coefficients for instantly dissolved nuclides.

5.2 Solute migration in and from repository

5.2.1 Overview

The solutes can migrate by advection i.e. with the flowing water and by molecular diffusion. Diffusion lets the solutes move in flowing as well as in stagnant water. Water that flows through the waste will carry with it nuclides at a concentration that depends on either the release rate from the waste form, e.g. by release from corroding metals, by dissolution of other solid phases that contain them or by desorption of them from the mineral phases on which they have sorbed. Nuclides that can immediately dissolve in the pore water of the waste will be carried out by the displacement of pore water by non-contaminated water. The flow rate of water through the waste will to a large degree determine the rate of nuclide release by advection. In addition molecular diffusion can cause considerable escape. Actually it can dominate over advection when low permeability barriers are present.

It is possible to set up and solve a system of equations that describe the water flowrate everywhere in the waste, buffer and rock including large and small fractures and fracture zones that intersect the vault. With the flowrates known the Advection-Diffusion/Dispersion for non-sorbing, sorbing and decaying nuclides can be solved. This can give information on the release rate of the nuclides to the water in the rock. There are several sophisticated programs that can be used to do this. The use of such programs needs considerable insights into the various governing processes and mechanisms. Also considerable experience in numerical analysis is needed to avoid some difficult to detect pitfalls. In the present problem there are several, sometimes very different characteristic times, that differ enormously between nuclides due to differences in half-lives and sorption coefficients. Very different space discretizations are needed for different nuclides to obtain correct results.

5.2.2 Simplified approach

We therefore use a simplified approach that is aimed at giving insights into the different processes. Sometimes very simple models and equations are derived for scoping calculations. These simple models can be used for screening purposes to see when some nuclides will not cause problems. Another important use of the simple models is to identify weaknesses and strengths in the different design options. A further use of the simple models is to show when and where a discretisation scheme in a sophisticated model should be given special attention. The latter is outside the scope of this work but has proven to be essential in some tests made with sophisticated tools.

When the flowrate through the waste is small the advective carrying capacity can be small compared to that by molecular diffusion. This will apply for conditions when low permeability barriers such as concrete, compacted clay or combinations thereof surround the waste. Then the low diffusivity in the barriers may set an upper bound on the rate of escape from the waste to the seeping water in the rock. The carrying capacity of the seeping water in the rock may limit the rate of nuclide escape.

In the diffusive mode the system can be visualised as having a series of transport resistances that all contribute to slow down the nuclide escape. The barrier with the largest resistance will set the pace for the escape rate of the nuclides as these resistances are coupled in series. This is analogous to an electrical circuit. Resistances in series and in parallel are also handled in the same way here when a barrier can have parallel paths.

When the carrying capacities by advective flow and by diffusion are similar in magnitude both processes contribute. The carrying capacity for advective flow is essentially the flowrate itself that flows through the waste. Similarly an equivalent flowrate, called Q-equivalent or Q_{eq} may be defined for the carrying capacity when diffusion dominates. In this way the importance of the two modes can be compared in a simple way.

5.2.3 Q-equivalent concept

This has recently been described in some detail for different conditions in Neretnieks et al. (2010). Here brief summaries of the concept together with some formulas that will be used later are presented.

The nuclide transport rate by the water *flowing through* the waste and buffer, N_Q is obtained by the product of the flowrate Q and the concentration c_{exit} in the water as it exits from the buffer

$$N_Q = Q c_{exit} \quad (5-1a)$$

In this case the flowing water out from the vault carries the nuclide. In addition, when there is molecular diffusion the nuclides can diffuse through the waste and the buffer without flow to reach the outside of the buffer and the seeping water in the rock. For this process the flowrate of a solute N_{Qeq} for steady-state conditions can be expressed by

$$N_{Qeq} = (c_{waste} - c_{water}) \frac{A}{\sum R_i} = Q_{eq,tot} (c_{waste} - c_{water}) \quad (5-1b)$$

A is the area through which transport occurs and R_i stands for the diffusion resistance in barrier “i”, which is coupled in series with other barriers. R_i for a barrier e.g. compacted clay is its thickness divided by the effective diffusion coefficient in it. c_{waste} is the concentration in the waste and c_{water} that in approaching water. This is equal to zero for practically all nuclides. One exception is radium, which is present in granitic groundwaters.

Equation (5-1b) is useful for nuclides with very long half-lives and little retardation, when steady-state conditions in the barriers prevail or at least are approached.

For nuclides with not very long half-lives the decay in the barriers must be accounted for specifically to obtain the concentration at the outside of the buffer c_{RI} . The RI stands for rock interface, which is taken to be the same as at the outside of the buffer. Equation 5-1b becomes

$$N_{Qeq} = Q_{eq} (c_{RI} - c_{water}) \quad (5-1c)$$

Q_{eq} is the same for all nuclides. The concentration at the rock interface c_{RI} will vary with time. It must be determined by solving the equations that govern the instationary diffusion in the barriers. This will be treated in Chapter 6. Equations (5-1a, b and c) are central to the later discussions and examples.

$$R_i = \frac{\Delta x_{buffer}}{D_{e,buffer}} \quad (5-2)$$

Individual Q_{eq} for a barrier i can also be defined by

$$Q_{eq,i} = A \frac{D_{e,i}}{\Delta x_i} \quad (5-3)$$

For the seeping water in the rock a simple expression can be derived by considering how far into the seeping water from the interface between the rock and buffer the solute has diffused during the time it is in contact with buffer, t_{res} , where the concentration at the rock interface is c_{RI} .

During this time the water has been into contact with the buffer, travelled along the interface and left it, the water has been exposed to the concentration c_{RI} at the buffer/rock interface. During the water passage the solute has diffused further and further out into the water. A concentration profile has built up. The situation can be approximated by diffusion into stagnant water exposed at one boundary to concentration c_{RI} during t_{res} . In the pore water in the rock the nuclide can diffuse a long distance before encountering any boundary.

Assuming smooth flow of water in the fractures in the rock a simple estimate of the mass transfer rate can be made. From diffusion theory we have that the mean penetration depth η_{mean} of the solute can be determined by integrating the concentration profile from the surface to infinity (Bird et al. 2002, p 621). That will be the distance from the surface of the buffer, which the solute has diffused from the buffer and will have attained the concentration c_{RI} . It is

$$\eta_{mean} = 1.13\sqrt{D_w t_{res}} = \frac{\int_0^{\infty} c dx}{c_{RI}} \quad (5-4)$$

D_w is the diffusion coefficient of the solute in water. It is different for different solutes but is typically in the range 1 and 3×10^{-9} m²/s for small molecules and ions in water at temperatures expected at repository depth. Considering the uncertainties in determining the water velocity and fracture aperture it is deemed permissible to assume that D_w is the same for all nuclides and equal to 10^{-9} m²/s according to Table 4-2.

The equivalent flowrate that has gained the concentration c_{RI} for flow perpendicular to the waste vault is

$$Q_{eq} = 2u\delta\eta_{mean} \cong 3.2\delta\sqrt{D_w r_h u} \quad (5-5)$$

u is the water velocity, $\delta = L \times \varepsilon$ is the width of the tunnel interface that is in contact with the flowing water. L is the length of the backfilled tunnel, r_h the tunnel radius and ε is flow porosity.

The factor 2 comes from the fact that fluid passes both sides of the tunnel. The above simple formula has neglected the curvature of the tunnel. The exact solution for flow perpendicular to the cylinder is (Neretnieks et al. 2010)

$$Q_{eqR,r} = 4.51\delta\sqrt{D_w r_h u} \quad (5-6)$$

This expression can also be used to calculate the equivalent flowrate for a fracture in the rock and a fracture zone intersecting the tunnel. However, for flow along the vault Equation (5-7) is more appropriate because the flow lines are straight. For flow along the tunnel the equivalent flowrate is

$$Q_{eq,L} = 1.13\delta\sqrt{D_w L u} \quad (5-7)$$

where $\delta = \pi 2r_h \times \varepsilon$ for a circular tunnel. When the gradient is at an angle α from the normal to the tunnel the equivalent flowrate can be approximated by

$$Q_{eq,\alpha} = Q_{eq,r} \times \sqrt{\cos(\alpha)} + Q_{eq,L} \times \sin(\alpha)$$

It may be noted that the Q_{eq} 's only depend on the properties of the rock and not on nuclide or buffer properties.

Main findings of Section 5.2.3

Equations were developed to calculate the equivalent flowrate for a vault. They have been adapted for use for a long repository vault with any flow direction in relation to the repository vault. These equations are central to the release calculations for all repository concepts.

5.2.4 Flowrate through vault

Flow through vault with gradient perpendicular to it

A bounding situation is when the hydraulic gradient is perpendicular to the vault. We neglect the end effects of the vault at present. Then flow in the vault filled with a highly conductive gravel will be flown through by very close to 2 times as much water as the same cross section of rock would have. This is shown in Figure 5-1. This and the next figure are based on the vault modelled as an elongated ellipsoid.

K_{in} is the conductivity in the concrete or bentonite in the vault and K_{out} that in the rock.

In the other extreme case when the vault is filled with bentonite the total flowrate will be close to 10 times less than in rock, (proportional to the clay conductivity). The presence of highly conductive waste inside the gravel or clay can strongly influence the flowrate through the vault. This is analysed in Section 6.2.

Flow through vault with gradient along the main axes

When the flow is along the vault the situation is quite different. The flowrate increases strongly but levels out at large conductivity ratios. The flowrate is much larger than for flow across the vault. Holmén (1997) estimated the flowrate to be about 100 times larger than if there is no vault, for vault dimensions similar to those used here. This can also be seen in Figure 5-2.

For K-ratios larger than 10 the relative flowrate increases considerably and reaches a maximum that depends on the axes ratio.

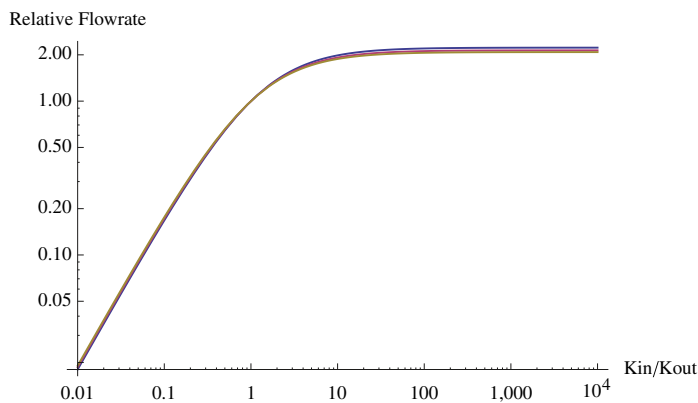


Figure 5-1. Flowrate in tunnel as function of the ratio of conductivity in tunnel to that in the rock. Model with ellipsoid from Carslaw and Jaeger (1959, p 427). Axes ratios 10, 16.7 and 40.

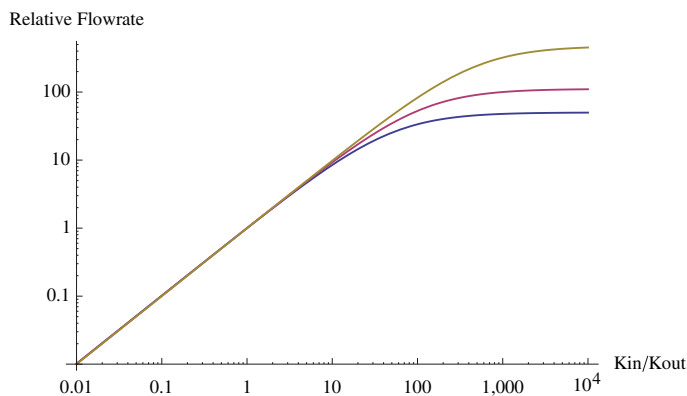


Figure 5-2. Flowrate in tunnel as function of the ratio of hydraulic conductivity in tunnel to that in the rock. Flow along tunnel. Model with ellipsoid from Carslaw and Jaeger (1959). Axes ratios 10, 16.7 and 40. From bottom and up.

5.2.5 Flowrate through vault with buffer

The relative flowrate inside the vault in the flow direction for flow across the vault is shown in Figure 5-1. It is based on the assumption that the conductivity of the buffer, K_{in} , including waste, is the same everywhere inside the vault. Figure 5-2 shows the relative flowrate for flow along the vault. The relative flowrate is the ratio between the flowrate in a vault with a buffer conductivity K_{in} , when rock has conductivity K_{out} . For equal conductivity the relative flowrate is one.

It is seen that when the buffer has lower conductivity than the rock the flowrate through the buffer decreases accordingly. For conductivity ratios larger than one the flowrate through the buffer increases to at most about a factor two for flow across the vault. For flow along the vault the flowrate can increase to very large values. This is further aggravated if the waste inside the buffer has a higher conductivity than the buffer. A considerable increase in flowrate through the waste will result if the *gradient is aligned with the vault*. If the gradient is perpendicular the increase in flowrate through the waste is relatively minor (i.e. a factor of 2 at most).

Main findings of Sections 5.2.4 and 5.2.5

When the waste and buffer has a *larger* conductivity than the rock very large flowrates of water can flow through the waste if the hydraulic gradient is more parallel to the vault than perpendicular to it. When the conductivity of *both* waste and buffer is *lower*, the flowrate through the waste is lower than through the same volume of rock. The case when the conductivity of the waste is larger than the buffer is more complex and is treated in Chapter 7. The nuclide release to the rock with the water seeping through the waste and buffer is proportional to the water flowrate and the concentration in the water leaving the buffer. That concentration will vary over time and be influenced by retardation and decay in waste and buffer. The release to the seeping water in the rock by molecular diffusion through waste and buffer is proportional to the equivalent flowrate and the concentration at the outside of the buffer i.e. at the buffer rock interface. Also this concentration is influenced by retardation and decay in the waste and buffer.

5.3 The hydraulic cage

5.3.1 Transport by diffusion from waste into flowing water in vault – application of the Qeq concept to hydraulic cage case

In this case the flowing water is exposed to nuclide at one boundary as previously, but the gravel extends only to the interface between gravel and rock. In the earlier derivation of the Q-equivalent model it was assumed that the nuclide could diffuse essentially an unlimited distance into the fractures in the rock. In the present case the nuclide diffuses in the large pores of the gravel up to its outer boundary. Thereafter it continues to diffuse into the fractures as in the previous case. The porosity of the gravel is vastly larger than that of the rock and the flow velocity correspondingly smaller. Here we wish to explore how the solute diffuses in the pores of the gravel and when this body of water could become equilibrated to the same concentration as at the surface of the waste.

Figure 5-3 shows how water seeping through and flowing around the waste becomes contaminated with nuclides. There are two different mechanisms that contribute to the leaching of nuclides from the waste. Water flowing through the waste becomes saturated to the solubility concentration or is determined by the dissolution rate of nuclides in the waste. However, in a good hydraulic cage very little flows *through* the waste. Water *passing* the waste is contaminated by diffusion from waste into the water. The contamination due to diffusion depends on the water residence time, the geometry of vault and on the water flow direction.

The nuclides in the waste enter the water flowing past it by diffusion from the interface between the waste and the flowing water. In addition the water that flows through the waste also carries dissolved nuclides. Although these entities are not strictly additive we approximate them to be additive. Two main cases are considered: the *constant solubility* case and the *constant dissolution rate* case. In the constant solubility case it is assumed that the nuclide concentration in the waste is set by the solubility of the nuclide. In the constant dissolution rate case it is assumed that the nuclide is released at a constant rate to the pore water in the waste. The concentration of a slowly decaying nuclide will reach a steady state determined by the balance of rate of release and the rate by which is carried out from the waste by flow and diffusion.

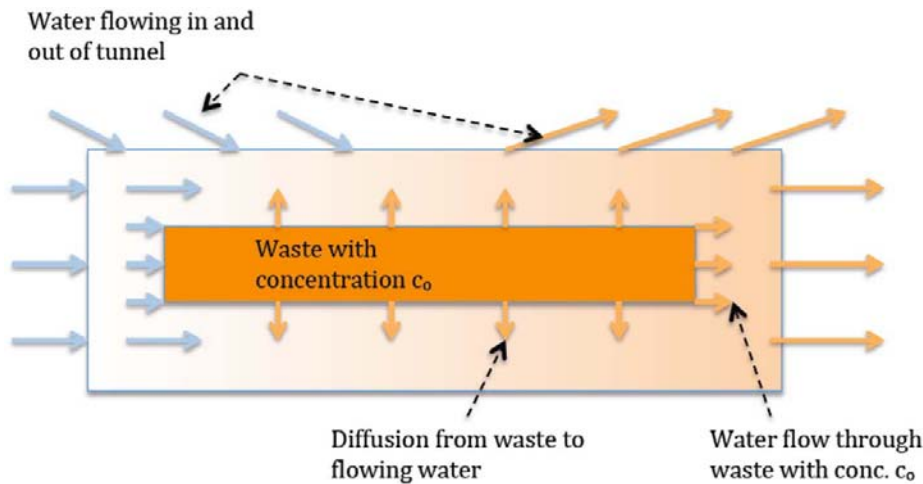


Figure 5-3. Illustration of how water flows into the vault from the rock and is contaminated by nuclides from the waste.

In the *constant solubility case* the release to and uptake in the flowing water is illustrated in Figure 5-4 where water flows in the gravel. The nuclide diffuses up into the water and by the time it leaves the vault the nuclide has attained a relatively high concentration near the waste and a lower concentration further out. If the water flows rapidly the nuclide will not have reached the outer boundary, the rock. If it flows slowly all the water may have attained the same concentration as that at the waste surface. In the *constant dissolution rate case* the dissolution rate itself will determine the release rate to the flowing water.

In the constant solubility case the release *by flow* through the waste is

$$N_{waste,flow} = Q_{waste,flow} c_0 \quad (5-8)$$

The release rate *by diffusion* to the water flowing past when *steady state conditions* prevail is

$$N_{diff} = Q_{eq} c_0 \quad (5-9)$$

where the equivalent flowrate Q_{eq} is approximated by (appendix in Neretnieks et al. 2010).

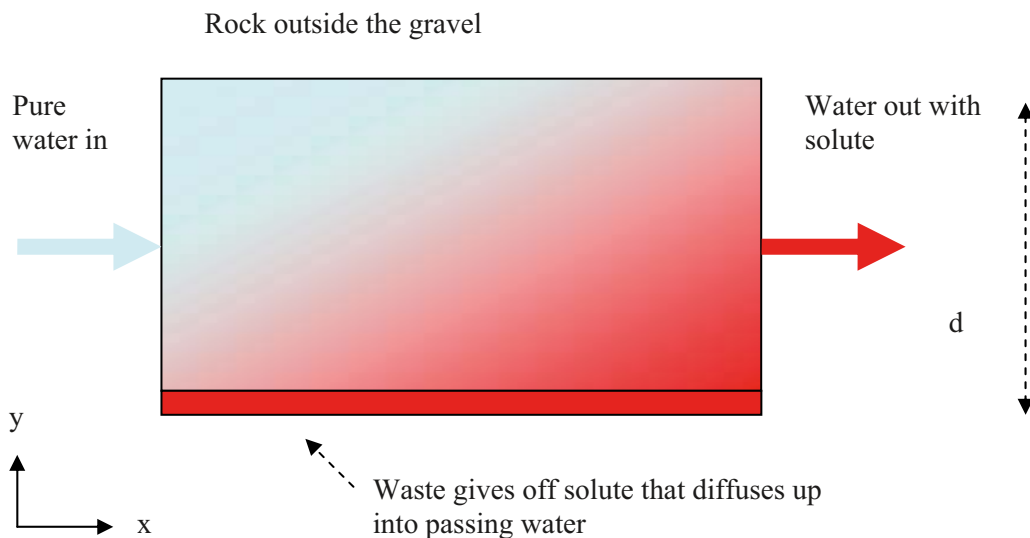


Figure 5-4. Water flowing through the gravel is contaminated by a solute released from the buffer surface.

$$Q_{eq} = Q_{flow} \left(1 - 2 \sum_{n=0}^{\infty} \frac{1}{\left(n + \frac{1}{2}\right)^2 \pi^2} e^{-\frac{\left(n + \frac{1}{2}\right)^2 \pi^2 D_a t_{res}}{d^2}} \right) \quad (5-10)$$

The apparent diffusivity coefficient D_a is

$$D_a = \frac{D_p}{R_a} \quad (5-11)$$

where R_a is the retardation factor

$$R_a = 1 + \frac{K_d \rho (1 - \varepsilon)}{\varepsilon} \quad (5-12)$$

and the nuclide residence time in the flowing water t_{res} is

$$t_{res} = t_w R_a \quad (5-13)$$

It may be noted that $D_a t_{res} = D_p t_w$ which implies that the Equation (5-10) is independent of sorption when steady conditions are reached. D_p is the pore diffusion coefficient and t_w the water residence time. One can also describe the process by determining which average concentration c_{mean} there is in the water that exits the vault. This is directly obtained from the expression below

$$c_{mean} = c_o \left(1 - 2 \sum_{n=0}^{\infty} \frac{1}{\left(n + \frac{1}{2}\right)^2 \pi^2} e^{-\frac{\left(n + \frac{1}{2}\right)^2 \pi^2 D_a t_{res}}{d^2}} \right) \quad (5-14)$$

Q_{flow} in Equation (5-10) is taken to be the largest flowrate (the flowrate varies along the vault) through the gravel.

Some sample calculations are presented below. From the calculations of flow in the ellipsoids, presented above, it was found that the total flowrates through the ellipsoid along the main axis and across it are 7.1 and 2.6 m³/yr respectively for very permeably gravel. The volume of the gravel in the idealised cylindrical tunnel case is 17,100 m³. The gravel filling has a porosity of 0.3. This gives mean water residence times 740 and 2,000 years for flow along and across the tunnel respectively.

The water flowing through the gravel is “rapidly” contaminated with the nuclides from the waste by diffusion. Figure 5-5 illustrates the how c_{mean}/c_o increases with water residence time in the gravel.

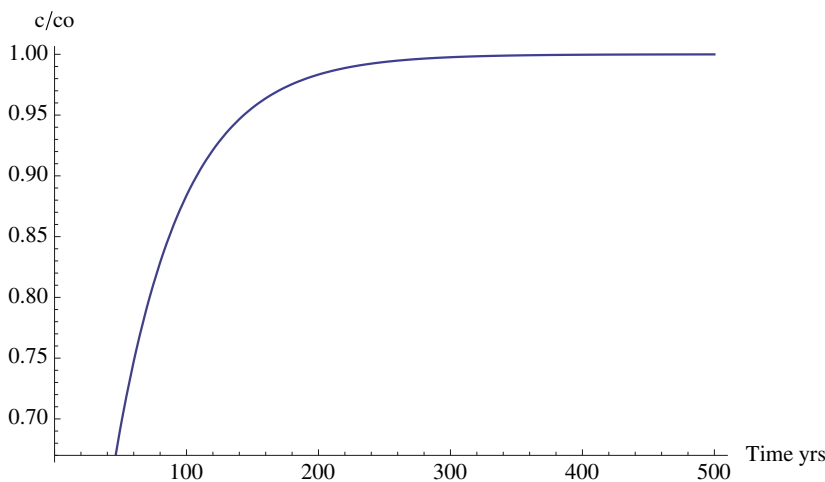


Figure 5-5. Effluent concentration from tunnel filled with gravel as function of residence time in the tunnel for a non-sorbing nuclide in the gravel when steady state conditions have been reached.

The time to reach steady state conditions can be approximately estimated from the time it takes to near-equilibrate all the water in the gravel provided the flow is so slow that the concentration is not swept away before this. The time to near-equilibrate the water can be estimated from the time constant for diffusion and is

$$t_{c,Diff} = \frac{d^2}{D_a} = \frac{d^2}{D_p} R_a \quad (5-15)$$

The time for flow to sweep it away is t_{res} .

When there is negligible sorption, which is assumed when the gravel consists of large particles $R_a = 1$. With the data in Tables 5-1 and 5-2 $t_{c,Diff} = 250$ years and t_{res} is 740 to 2,000 years depending on the direction of the gradient. Steady state conditions are therefore reached in about 250 years because the concentration profile develops in much shorter time than the water sweeps away the water. All water is fully equilibrated. It may be noted that for sorbing nuclides the ratio of $t_{c,Diff}/t_{res}$ is the same as both times are increased by the retardation factor.

The constant dissolution rate case is not further treated because it is evident from the above treatment that if it takes on the order a few hundred years for a non-sorbing nuclide to pass the gravel, the gravel case is not an efficient barrier in whatever manner the release from the waste occurs.

Main findings of Section 5.3

The analysis shows that the water that is drawn in through the gravel in a hydraulic cage will be fully equilibrated with the nuclides in the waste. It is as if all the water has flown through the waste. The hydraulic cage is not a good option.

5.4 Equivalent flowrates in fractures and fracture zones intersecting the tunnel

It is assumed that fracture zones and fractures with large transmissivities cannot be entirely avoided. Equation (5-6) can be used to calculate Q_{eq} for such features in cases where the hydraulic conductivity of the buffer is low. For a single fracture δ is the fracture aperture. For a fracture zone L wide and with the flow porosity ϵ of the zone, $\delta = L \times \epsilon$ when the fracture intersects the tunnel perpendicularly. If the feature intersects the tunnel at an angle α from that normal to the tunnel axis the radius can approximately be taken to be to $r_h / \cos(\alpha)$. This is not valid when α approaches $\frac{\pi}{2}$, i.e. when the zone intersects the tunnel along its entire length. A tunnel will probably not be excavated along a fracture zone but a long fracture might still be present. In this case the tunnel length is used. For the parallel case the results shown in Table 5-1 are obtained for one prominent fracture and one fracture zone with Equation (5-6). The table also includes results for a fracture zone that intercepts the whole length of the tunnel to illustrate the consequences.

Table 5-1 summarises the data used in the sample calculations. For comparison the equivalent flowrate for the whole tunnel surrounded by rock is also shown. We remind the reader that these results apply to cases where the buffer has a much lower conductivity than the rock.

Table 5-1. Q_{eq} for a fracture and a fracture zone with the hydraulic gradient perpendicular as well as parallel to the tunnel.

Perpendicular to tunnel	T m ² /s	K m/s	ϵ	L m	δ m	u m/yr	Q_{eq} m ³ /yr
Rock		3.00E-09	1.00E-04	200	2.00E-02	2.8	0.0747
Single fracture	1.00E-06		1	1.00E-03	1.00E-03	95	0.0037
Fracture zone		1.00E-06	0.01	2	2.00E-02	9.5	0.1365
Parallel to tunnel							
Rock		3.00E-09	1.00E-04	200	4.80E-03	2.8	0.0228
Single fracture	1.00E-06		1	200	2.00E-03	95	0.0553

The Q_{eq} for the rock decreases slightly more than three times when the flow angle changes from perpendicular to parallel. It will lie between these values for other angles. The same will apply to the single prominent fracture. It is seen that a fracture zone has nearly twice the equivalent flowrate than the whole tunnel. A single prominent fracture has a 20 times lower equivalent flowrate than the whole tunnel in the "perpendicular to tunnel" case.

Main findings of Section 5.4

The presence of a few prominent fractures does not drastically increase the equivalent flowrate, above that for the base case fractured rock, when the buffer and waste has a *lower* conductivity than the rock. A fracture zone of the type in the example increases the equivalent flowrate noticeably.

5.5 Flowrate through very permeable buffer and waste intersected by a fracture

A "what if" case is explored where it is assumed that the buffer (clay or concrete) has been severely fractured or degraded. The waste is also assumed to have a high conductivity and to exert negligible resistance to flow. The vault is intersected by a prominent fracture ($T = 10^{-6} \text{ m}^2/\text{s}$) or a fracture zone. The transmissivity of the fracture sets the limit to the flowrate. With the data in Table 5-1 for *one* fracture perpendicular to the vault the flowrate through the waste is obtained from

$$Q = 2 \times 2r_h \times \delta \times u \quad (5-16)$$

For the prominent fracture the flowrate through the waste is 2.9 m³/yr. For the fracture zone it is 5.8 m³/yr.

Main findings of Section 5.5

The presence of a few prominent fractures does not drastically increase the equivalent flowrate, above that for the base case fractured rock, when the buffer and waste has a *lower* conductivity than the rock. A fracture zone of the type in the example increases the equivalent flowrate noticeably.

5.6 Summary of flowrates and equivalent flowrates for base case conditions

Table 5-2 summarises flowrates and equivalent flowrates for different conditions for the base case. The flowrates through the vault with different infill (undisturbed rock, gravel, concrete and clay) are calculated approximating the vault by an ellipsoid as described earlier. Two flow directions are considered. In one case the gradient is along the vault in the other it is perpendicular to it.

Some comments to the table are given below.

Row 1 shows the flowrate through the vault volume before rock excavation for a hydraulic gradient of 0.003. When the gradient is perpendicular to the vault the flowrate is more than 15 times as large as when flow is parallel to it. This is because the cross section area is 15 times larger for flow across the vault.

Rows 2 and 3 gives the maximum flowrates in the vault when there is no flow resistance (empty vault) and when it is filled with gravel with much lower resistance than the rock. It may seem surprising that the flowrate for flow along the vault increases so much when it increases only a factor two for perpendicular flow. For parallel flow the water is drawn from a large region as illustrated in Figure 5-6. This will be further explored in Chapter 7.

Rows 4 and 5 gives the maximum flowrates when the vault is fully filled with concrete and clay respectively. The flowrates decrease about proportionally to the decrease of the conductivity of the infill compared to the case of unexcavated vault (Row 1).

Rows 6 and 7 give the flowrates that would result if the bentonite or concrete were very degraded at least locally where the vault is intersected by the prominent fracture or fracture zone

Rows 8 to 10 show *equivalent flowrates*. Row 8 gives the value for the seeping water in the fractured rock. When there is negligible flow through the repository as for row 4 where the vault is filled with concrete that has retained its low hydraulic conductivity, this equivalent flowrate sets the ultimate carrying capacity of nuclides from the repository. When clay fills the vault, row 5, it allows some water to flow through the repository with a magnitude that is similar to the equivalent flowrate, row 8. As a first approximation one could add the equivalent flowrate and the flowrate of water through the repository to obtain the total carrying capacity. Q_{eq} in the rock is much smaller than Q_{eq} in clay and concrete and will be rate determining for the release by diffusion.

Rows 9 and 10 give the equivalent flowrates for diffusion through concrete and clay. These resistances ($1/Q_{eq}$) are in series with and are much smaller than the resistance to transport in the rock, which implies that they will contribute little to the over-all resistance for a non-decaying or very long-lived nuclide.

When the resistances in series $\frac{1}{Q_{eq,clay}}$ and $\frac{1}{Q_{eq,rock}}$ are added the total equivalent flowrate $Q_{eq,tot}$ becomes, for e.g. for the clay case

$$Q_{eq,tot} = \frac{1}{\frac{1}{Q_{eq,clay}} + \frac{1}{Q_{eq,rock}}} \cong Q_{eq,rock} \quad (5-17)$$

because $Q_{eq,clay} \gg Q_{eq,rock}$

Table 5-2. Flowrates and equivalent flowrates for different conditions for the base case.

	Q _{eq} or Q m ³ /yr		Comments
	Flow parallel to tunnel	Flow perpendicular to tunnel	
Flowrates			
1 Flowrate through tunnel volume before excavation, Q	0.072	1.2	Flowrate through the intact rock volume where the tunnel will be located
2 Flowrate through empty tunnel, Q	8.0	2.6	Flowrate after resaturation
3 Flowrate through tunnel filled with gravel, Q	7.1	2.6	K _{gravel} = 3000*K _{rock} = 1E-6. Flowrate after resaturation
4 Flowrate through tunnel filled with concrete, Q	1.20E-04	3.70E-03	K _{concrete} = 5E-12 m/s. Flowrate after resaturation
5 Flowrate through tunnel filled with clay, Q	2.40E-03	0.069	K _{clay} = 1E-10 m/s. Flowrate after resaturation
6 Flowrate via prominent fracture when buffer is very degraded, Q		2.9	T = 1E-6 m ² /s
7 Flowrate via fracture zone when buffer is very degraded, Q		5.8	T = 2E-6 m ² /s
Equivalent flowrates			
8 Equivalent flowrate in rock, Q _{eq}	0.023	0.075	K _{waste} and K _{buffer} << K _{rock} . Q _{eq} for whole tunnel
9 Equivalent flowrate in concrete, Q _{eq} , from diffusion resistance	1.34	1.34	D _e = 1E-11 m ² /s, Q _{eq} for whole tunnel
10 Equivalent flowrate in clay, Q _{eq} , from diffusion resistance	13.4	13.4	D _e = 1E-10 m ² /s, Q _{eq} for whole tunnel

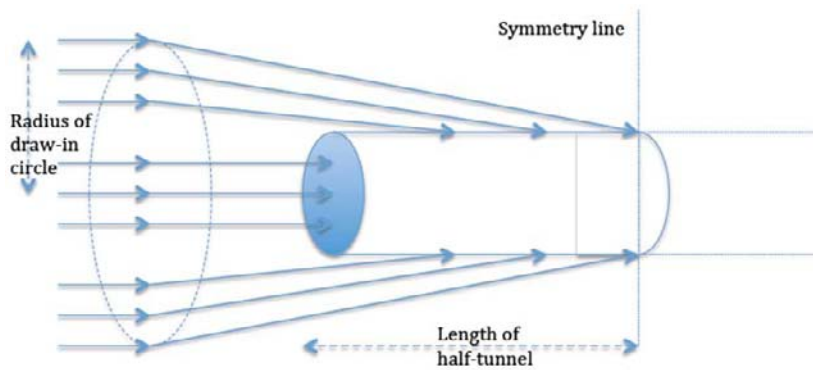


Figure 5-6. When the flow is along the vault with a medium with much larger conductivity than the rock much water is drawn into it.

In the previous descriptions of how water flowrates and molecular diffusion in waste, buffer and rock affect the nuclide release to the seeping water in the rock and with the data presented in Table 5-2 it is found that:

- In vaults with gravel (hydraulic cage) a large flowrate will be drawn into the vault.
- For a hydraulic cage the water that flows through the gravel will to a small extent flow through the waste but it will be fully contaminated by molecular diffusion.
- The release from the waste of the nuclides is assumed to be faster than the transport through either concrete or clay barriers, unless the waste is specifically designed to have low hydraulic conductivity and diffusivity.
- The flowrate in a vault filled with a buffering barrier (concrete or clay), with hydraulic conductivity markedly smaller than the surrounding rock, will be smaller than the flowrate through the unexcavated vault roughly in proportion to the ratio of hydraulic conductivity of buffer to rock.
- When the water flowrate through the (buffer in the) vault is much less than the equivalent flowrate, Q_{eq} , in the rock, the former can be neglected and the release to seeping water will be determined by Q_{eq} and the nuclide concentration in pore water at the interface between buffer and rock c_{RI} .

The use of flowrates and equivalent flowrates to compare carrying capacities can be used for steady state conditions of concentrations in the barriers. They contain no information on how the concentration of sorbing and decaying nuclides develop over time in the concrete and clay barriers. Steady state conditions can be expected to develop for non-sorbing nuclides with very long half-lives such as Cl-36 and I-129. For other nuclides the concentration at the outside of the buffer can deviate considerably from that in the waste inside the concrete and clay barrier because it will take time for the nuclide to diffuse through the barrier. Especially nuclides with short half-lives and strongly sorbing nuclides can decay considerably before they reach the outside of the barrier. For such nuclides one needs to assess the nuclide concentration at the outside of the buffer, c_{RI} . This is analysed in the next chapter.

Main findings of Chapter 5

A hydraulic cage will draw in large amounts of water, on the order of several m^3/yr . All this water will be contaminated. The same applies if the buffer should become severely damaged to attain conductivity larger than the rock. Under such conditions a prominent fracture or fracture zone can further increase the flowrate. This water can be considerably contaminated with nuclides. An intact concrete in buffer *and* waste limits the flowrate *through* the concrete and waste to a few litres/yr. Intact clay with 20 times larger conductivity than concrete will allow 20 times larger flow of water. The nuclides that diffuse out through the buffer into the seeping water in the rock will contaminate an equivalent flowrate of some tens of litres/yr. For *steady state conditions* the diffusion resistance for non-decaying nuclides in the concrete or clay is small compared to that in the rock and will not significantly aid in decreasing the overall equivalent flowrate.

6 Diffusion dominated transport in bentonite and concrete

6.1 Diffusion and decay in the buffer

When there is little or no flow through the bentonite or concrete the nuclides escape by diffusion only. In the several metres thick barrier the diffusion of sorbing nuclides is retarded by sorption. First some simple cases are described and applied to the suite of nuclides of interest. For these cases simple analytical solutions can be used. In one case considered, the nuclide concentration c_o at the boundary between the waste and the barrier is constant. This could be the case when the waste form is dissolving “rapidly” but the released nuclide cannot fully dissolve at the rate at which it is released, because of solubility limitations. In another case the decaying influx $N_o e^{-\lambda t}$ into the barrier is used as boundary condition. This could be the case for a case where the waste *matrix dissolves at a constant rate* releasing the nuclide, which, however, due the radioactive decay has a decreasing concentration over time. The *nuclide release rate decays exponentially* with time. A third case is when the nuclide has dissolved fully in the waste porewater at an early stage. Neglecting the loss by escape, the concentration in the waste is then $c = c_o e^{-\lambda t}$. It is found that for all three cases a single parameter group, $H = x_o \sqrt{\frac{\lambda R_a}{D_p}}$, can be used to assess when a nuclide will decay to insignificant values before emerging at the outside of a barrier. This is used to compare barriers with different sorption and diffusion properties.

Analytical solutions of the diffusion equation for these cases can be found in Neretnieks (1984). The concentration at a location x_o in an infinitely thick barrier or the flux past x_o in the barrier can be calculated. The infinitely thick barrier is used in the modelling because this gives very simple expressions. It was shown that for small out-fluxes or low concentrations at x_o the error caused by using an infinite medium instead of one with a thickness x_o is small and acceptable for our purposes.

The solution for decaying *inflow rate* at the inlet is

$$N_{x_o} = N_o e^{-\tau} \text{Erfc}\left(\frac{H}{2\sqrt{\tau}}\right) \quad (6-1)$$

Erfc stands for the mathematical so called ”complimentary error function”. Similarly for a *decaying concentration* at the inlet boundary

$$c_{x_o} = c_o e^{-\tau} \text{Erfc}\left(\frac{H}{2\sqrt{\tau}}\right) \quad (6-2)$$

where

$$\tau = t\lambda \quad (6-3)$$

$$H = x_o \sqrt{\frac{\lambda}{D_a}} = x_o \sqrt{\frac{\lambda R_a}{D_p}} \quad (6-4)$$

For the *constant concentration* (solubility limitation) case the solution is

$$c_{x_o} = \frac{c_o}{2} \left(e^{-H} \text{Erfc}\left(\frac{H}{2\sqrt{\tau}} - \sqrt{\tau}\right) + e^H \text{Erfc}\left(\frac{H}{2\sqrt{\tau}} + \sqrt{\tau}\right) \right) \quad (6-5)$$

Figure 6-1 shows the results obtained from Equations 6-1, 6-2 and 6-5.

For very long times this approaches

$$c_{x_o} = c_o e^{-H} \quad (6-6)$$

When the buffer is in direct contact with the rock this is also the concentration at the rock interface c_{Rl} . For the decaying inflow rate case the outflow reaches a maximum at some time at x_o , whereas for the constant concentration case the concentration increases to a maximum value at very long times. Figure 6-2 shows the peak outflow for the inflow case and the constant concentration case (solubility limit) at long times.

It may be noted that both the outflow and the concentration drops very strongly for large H . For H larger than 20 the nuclide flowrate and concentration in most cases will have changed by a factor nearly 10^{-9} . Before steady state conditions are reached the concentration at x_o is even lower as can be seen in Figure 6-3.

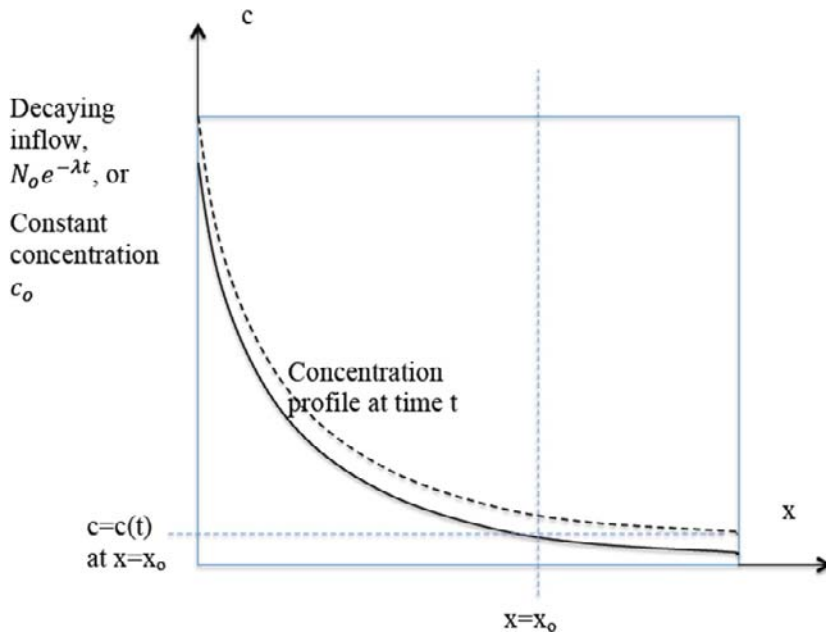


Figure 6-1. Illustration of concentration profile in an infinite barrier at time t . At the left boundary there is either decaying influx of nuclide (solid line) or constant concentration (dashed line).

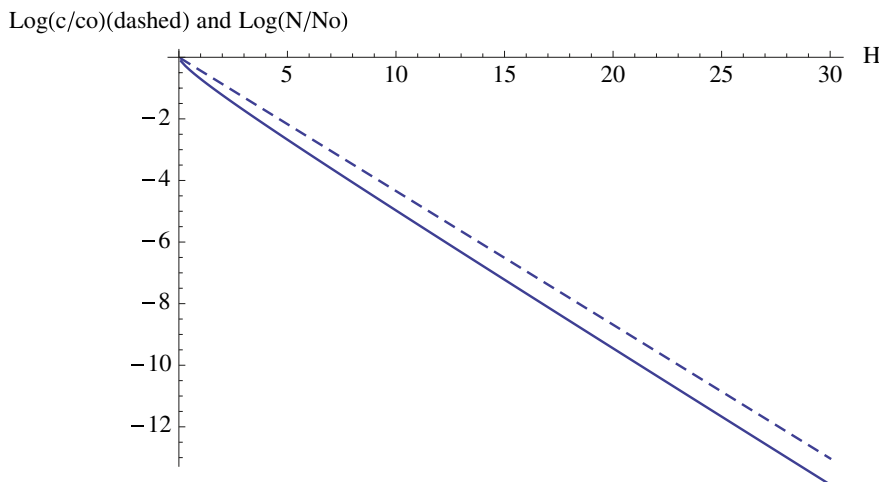


Figure 6-2. Maximum outflow and maximum concentration (dashed) as function of H .

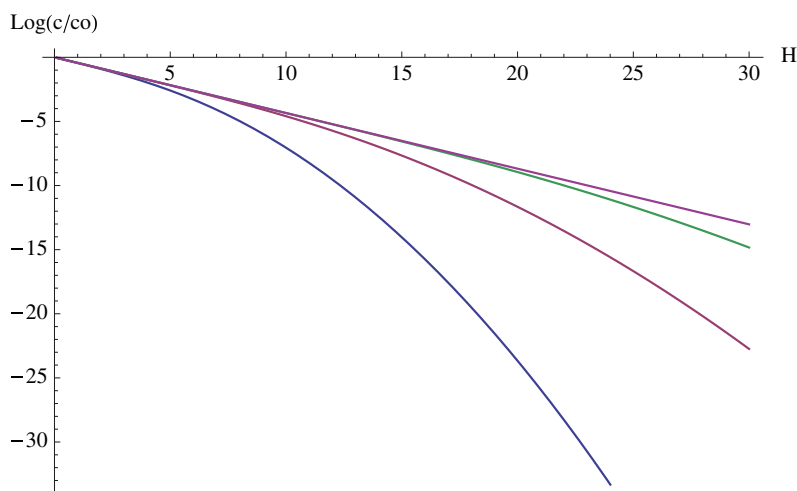


Figure 6-3. Concentration as function of H for $\tau = 2, 5, 10$ and 100 (highest curve).

Main findings of Section 6.1

A very simple tool is presented that can be used to assess how the thickness and diffusion and sorption properties of a buffer affect the release of nuclides from the barrier. This applies to buffers with low conductivity and negligible flow through them. It can be used to compare clay and bentonite barriers and to get a fast impression of the retention capabilities of the buffer. The model also shows how the thickness, diffusion and retardation properties of the concrete and clay impact on its retaining properties for different nuclides. Bentonite and especially concrete cause many sorbing nuclide decay to insignificance during their passage through the buffer. The simple formula Equation 6-6 is conservative because it assumes that steady state conditions have been reached.

6.2 Sample calculations

Table 6-1 shows the H -values for a number of nuclides passing through clay and concrete buffers. Results for non-degraded concrete as well as a fractured/degraded barrier are presented. A large H -value indicates large decay. For $H > 20$ the nuclide decays by a factor 10^9 during its passage through the barrier when steady state has been reached. The concrete, even when partly degraded, has larger H -values than clay because of the larger sorption coefficients and lower diffusion coefficients. However, the H -value alone does not say when the maximum release value is reached. Long-lived and strongly sorbing nuclides will reach steady state after many millions of years. A supplementary criterion that gives what maximum concentration could be reached after a given time e.g. 10^5 years is useful. It uses the ratio of the time constant for diffusion, $t_D = x_0^2/D_p$ discussed more in a later section, to the time of interest, in this case on the order of 100,000 years. If the ratio is larger than 20 the concentration at the outside of the buffer builds up to at most $1.5 \times 10^{-3}c_0$ at the end of the time. For a ratio of 40, the maximum concentration for a stable nuclide will be less than $10^{-5}c_0$ at the end of the period. This is valid for very long-lived or stable nuclides. Values of H and $t_D/10^5$ are given for a large number of nuclides in Table 6-1 for concrete as well as for bentonite buffers.

Main findings of Section 6.2

If the concrete and the clay barriers retain their low conductivity over long times and the waste also is not more conductive to flow than the buffer, the concrete barrier is superior to the bentonite in the extent to which nuclides can decay during their passage. Many important nuclides will decay considerably during their passage through especially the concrete buffer.

Table 6-1. Calculated values of H , t_D and R_a for 2 m thick concrete and bentonite barriers. Sorption data (Kd) are taken from Cronstrand (2005) using his recommended values. Concrete and bentonite porosities are 0.15 and 0.4.

Nuclide	Half-life yrs	Total activity Bq	Concrete $D_p = 6.7E-11$ H	Concrete $D_p = 6.7E-10$ H	Clay $D_p = 2.5E-10$ H	Concrete $D_p = 6.7E-11$ R_a	Clay $D_p = 2.5E-10$ R_a	Concrete $D_p = 6.7E-11$ $t_D/1E5$	Clay $D_p = 2.5E-10$ $t_D/1E5$
H-3	12	2.50E+15	10	1	5	1	1	0.0	0.0
Be-10	1.60E+06	1.40E+11	0	0	0	1	1	0.0	0.0
C-14	5.70E+03	2.00E+14	26	3	0	2,948	1	55.9	0.0
Cl-36	3.00E+05	2.70E+11	0	0	0	1	1	0.0	0.0
K-40	1.30E+09	1.00E+09	0	0	0	1	1	0.0	0.0
Fe-55	2.7	1.00E+15	22	2	11	1	1	0.0	0.0
Co-60	5.3	8.50E+15	382	38	72	590	79	11.2	0.4
Ni-59	7.50E+04	1.50E+15	3	0	1	590	79	11.2	0.4
Ni-63	96	1.40E+17	90	9	17	590	79	11.2	0.4
Se-79	6.50E+04	5.10E+08	1	0	0	89	1	1.7	0.0
Sr-90	29	3.10E+12	27	3	8	16	5	0.3	0.0
Zr-93	1.50E+06	2.30E+12	3	0	0	7,368	196	139.6	1.0
Nb-93m	14	8.60E+13	831	83	5	7,368	1	139.6	0.0
Nb-94	2.00E+04	5.10E+12	22	2	0	7,368	1	139.6	0.0
Mo-93	3.50E+03	2.00E+12	6	1	0	89	1	1.7	0.0
Tc-99	2.10E+05	9.10E+11	7	1	0	7,368	40	139.6	0.2
Pd-107	6.50E+06	1.30E+08	0	0	0	590	1	11.2	0.0
Ag-108m	1.30E+02	1.20E+12	13	1	2	16	1	0.3	0.0
Cd-113m	14	8.20E+09	235	24	45	590	79	11.2	0.4
Sn-126	1.00E+05	6.30E+07	10	1	0	7,368	40	139.6	0.2
Sb-125	2.8	3.30E+12	22	2	11	1	1	0.0	0.0
I-129	1.60E+07	3.80E+07	0	0	0	45	1	0.9	0.0
Cs-134	2.1	1.60E+12	99	10	59	16	21	0.3	0.1
Cs-135	2.30E+06	6.30E+08	0	0	0	16	21	0.3	0.1
Cs-137	30	4.20E+13	26	3	16	16	21	0.3	0.1
Ba-133	11	3.00E+10	11	1	6	1	1	0.0	0.0
Pm-147	2,6	1.50E+12	22	2	12	1	1	0.0	0.0
Sm-151	90	2.60E+11	1,037	104	55	73,668	781	1,396.2	4.0
Eu-152	13	1.50E+12	2,728	273	145	73,668	781	1,396.2	4.0
Eu-154	8.8	7.60E+11	3,316	332	177	73,668	781	1,396.2	4.0
Eu-155	5.0	1.80E+11	4,399	440	234	73,668	781	1,396.2	4.0
Ho-166m	1.20E+03	8.40E+10	284	28	15	73,668	781	1,396.2	4.0
Pb-210	22	2.70E+11	8	1	4	1	1	0.0	0.0
Ra-226	1.60E+03	3.80E+11	25	2	2	738	21	14.0	0.1
Ac-227	22	1.40E+06	8	1	4	1	1	0.0	0.0
Th-229	7.30E+03	1.40E+02	115	12	1	73,668	40	1,396.2	0.2
Th-230	7.70E+04	1.80E+05	35	4	0	73,668	40	1,396.2	0.2
Th-232	1.40E+10	1.10E+10	0	0	0	73,668	40	1,396.2	0.2
Pa-231	3.30E+04	2.50E+06	0	0	0	1	1	0.0	0.0
U-232	72	4.40E+06	1,159	116	14	73,668	40	1,396.2	0.2
U-233	1.60E+05	3.10E+04	25	2	0	73,668	40	1,396.2	0.2
U-234	2.40E+05	7.80E+08	20	2	0	73,668	40	1,396.2	0.2
U-235	7.00E+08	6.40E+09	0	0	0	73,668	40	1,396.2	0.2
U-236	2.30E+07	8.10E+07	2	0	0	73,668	40	1,396.2	0.2
U-238	4.50E+09	4.60E+10	0	0	0	73,668	40	1,396.2	0.2
Np-237	2.10E+06	1.80E+08	7	1	0	73,668	391	1,396.2	2.0
Pu-238	88	3.70E+11	1,049	105	125	73,668	3,901	1,396.2	19.8
Pu-239	2.40E+04	2.30E+12	63	6	8	73,668	3,901	1,396.2	19.8
Pu-240	6.50E+03	1.80E+12	122	12	15	73,668	3,901	1,396.2	19.8
Pu-241	14	4.40E+12	2,629	263	313	73,668	3,901	1,396.2	19.8
Pu-242	3.80E+05	1.30E+09	16	2	2	73,668	3,901	1,396.2	19.8
Pu-244	8.30E+07	1.80E+02	1	0	0	73,668	3,901	1,396.2	19.8
Am-241	4.30E+02	5.00E+12	212	21	57	14,734	3,901	279.3	19.8
Am-242m	1.50E+02	2.00E+09	359	36	96	14,734	3,901	279.3	19.8
Am-243	7.40E+03	8.50E+09	51	5	14	14,734	3,901	279.3	19.8
Cm-243	29	1.70E+09	817	82	218	14,734	3,901	279.3	19.8
Cm-244	18	4.40E+10	1,037	104	276	14,734	3,901	279.3	19.8
Cm-245	8.50E+03	7.70E+07	48	5	13	14,734	3,901	279.3	19.8
Cm-246	4.70E+03	2.10E+07	64	6	17	14,734	3,901	279.3	19.8

6.3 Simplified approach – release only by equivalent flowrate Q_{eq}

A simple way to assess an upper bound of the release of nuclide into the seeping water in the fractures in the rock is presented below. The Q_{eq} concept is based on release by only diffusion through the barriers to the seeping water in the fractured or porous rock. When the concentration at the interface between buffer and rock, c_{RI} is known the release rate $N_{Q_{eq}}$ to the seeping water in the rock around the whole repository is

$$N_{Q_{eq}} = Q_{eq} c_{RI} \quad (6-7)$$

6.3.1 Example

An upper bound of c_{RI} is obtained from Equations (6-4) and (6-6). This gives the concentration if no nuclide is swept away by the seeping water at the buffer/rock interface. As some is swept away the concentration will be lower. The error decreases with increasing H . With base case data for Ni-59 and a concrete buffer (See Table 4-2), $H = 3.21$ the relative concentration at the interface c_{RI}/c_o is 0.040.

In Section 6.1 the maximum nuclide concentration (in relative terms) at the buffer rock interface was calculated. These need to first be translated into absolute concentrations in the pore waters and then to release rates of nuclide with the equivalent flowrate Q_{eq} .

Non-sorbing and non-solubility limited nuclides are all dissolved in the pore water of the waste. For sorbing nuclides some will be sorbed and the pore water concentration will be less by the retardation factor

$$R_a = 1 + \frac{1-\varepsilon}{\varepsilon} K_d \rho \quad (6-8)$$

so that

$$c_o = \frac{A_{RN}}{R_a \times V_{Pore,waste}} \quad (6-9)$$

A_{RN} is the activity inventory and $V_{Pore,waste}$ is the water volume in the waste. For a waste porosity of 0.2 the water volume is 4,000 m³ for the geometry in the base case, for Ni-59

$$c_o = \frac{1.5 \times 10^{15}}{4000 \times 590} = 6.4 \times 10^8 \text{ Bq/m}^3 \text{ pore water}$$

In the example for Ni-59 the total inventory of nuclide is 1.5×10^{15} Bq. $Q_{eq} = 0.075$ m³/yr for the whole vault was used in the example (From Table 5-1, flow perpendicular to vault).

$$N_{Q_{eq}} = Q_{eq} R \times c_o \times \frac{c_{RI}}{c_o} = 0.075 \times 6.4 \times 10^8 \times 0.040 = 1.9 \times 10^6 \text{ Bq/yr}$$

Main findings of Section 6.3

This very simple method gives an upper bounding value of the release. It gives conservative results and can be used for screening purposes. It will later be shown that the rate of diffusion through the intact buffer of a nuclide is faster than by flow through the buffer. Under such conditions the rate of release of a nuclide to the water in the rock can, as a first approximation, be taken as the product of the equivalent flowrate from Section 5.6 and the nuclide concentration at the outside of the buffer determined by the simple formula in Section 6.1.

6.4 Diffusion in waste and buffer, decay and uptake by Q_{eq}

The method described above can be used for screening purposes, although it will tend to overestimate the release. In this section we explore how large the overestimation could be. For this purpose, a more detailed approach is developed that considers cases where the decay, release of and migration in the waste form itself also are accounted for. The waste is modelled as being a homogeneous

porous material where the nuclides are dissolved and where the sorbing species also are partly sorbed in the waste matrix. The cylindrical waste body in the base case has a radius r_v of 5.64 m. The thickness of the concrete or clay barrier surrounding the waste is 2 m. Thus, from the interior, the nuclides must diffuse a nearly three times longer distance in the waste before they reach the barrier to thereafter diffuse through the concrete or clay buffer. The simplified model also overestimates releases because it overestimates the concentration at the outer boundary of the buffer, i.e. the maximum c_{RI}/c_o , since it does not account for nuclides being flushed away by advection there.

Below a model is presented where the effect of adsorption in the waste and diffusion out of it retards the migration from the waste to the waste/buffer interface followed by migration through the buffer to the rock interface where seeping water in the rock carries it away. The migration in the waste and buffer is by diffusion only. The release to the seeping water is modelled by the Qeq approach.

The instationary diffusion equation (Fick's second law) is solved for this case for radial cylindrical symmetry. The initial condition is that in the waste there is sorption equilibrium so the concentration is given by Equation (6-9). In the buffer there is initially no nuclide. The nuclide that reaches the buffer/rock interface is carried away by the equivalent flowrate Q_{eq} . The inner boundary condition for the waste is taken as the symmetry condition in the middle of the waste. The following equations are solved:

$$R_a(r) \frac{\partial c_p}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(D_p(r) r \frac{\partial c_p}{\partial r} \right) - \lambda R_a(r) c_p \quad (6-10)$$

The retardation factors as well as the pore diffusion coefficients are different in the waste and buffer. The initial condition is

$$c_p = c_o \text{ for } t = 0 \text{ and } 0 < r < r_v \text{ and } c_p = 0 \text{ for } r > r_v \quad (6-11)$$

The boundary conditions are

$$\frac{\partial c_p}{\partial r} = 0 \text{ for } r \rightarrow 0 \quad (6-12)$$

and

$$D_e A \frac{\partial c_p}{\partial r} = -Q_{eq} c_p \text{ at } r = r_b \quad (6-13)$$

r_v and r_b are the radii of waste and buffer respectively. The pore diffusion coefficient D_p and retardation factor R_a jump from one value in the waste to another value in the buffer. The equations are solved numerically by the method of lines using Mathematica®.

6.5 Sample calculations

6.5.1 Cl-36

Cl-36 has a half-life of 3×10^5 years and does not sorb on waste, concrete or bentonite. It is therefore expected that Q_{eq} and the concentration in the pore water in the waste will determine its release to the seeping water in the rock. For flow perpendicular to the vault $Q_{eq} = 0.075 \text{ m}^3/\text{yr}$. With inventory data from Table 6-1, $2.7 \times 10^{11} \text{ Bq}$, the pore water concentration $c_o = 6.8 \times 10^7 \text{ Bq/m}^3$. The release using $c_o \times Q_{eq}$ is $5.1 \times 10^6 \text{ Bq/yr}$. The numerical solution gives $2.6 \times 10^6 \text{ Bq/yr}$ for perpendicular flow. As expected the release estimated by $c_o \times Q_{eq}$ is higher than that obtained by the numeric solution because the latter accounts for diffusion in and for decrease of the concentration in waste as the nuclide is gradually washed out.

6.5.2 Ni-59

For concrete as buffer with the base case data, Figure 6-4 shows the relative concentration in the pore water in waste and buffer (fraction of starting concentration in waste c_o) over time. Ni-59 has a half-life of 75,000 years. The parameter $H = 3.21$ and the maximum concentration at the rock interface calculated using Eq. 6-6 is $0.040 c_o$ compared with $0.023 c_o$ by the numerical solution. The relative pore water concentration c_p is the ratio of the concentration on the outside of the buffer to that on the inside.

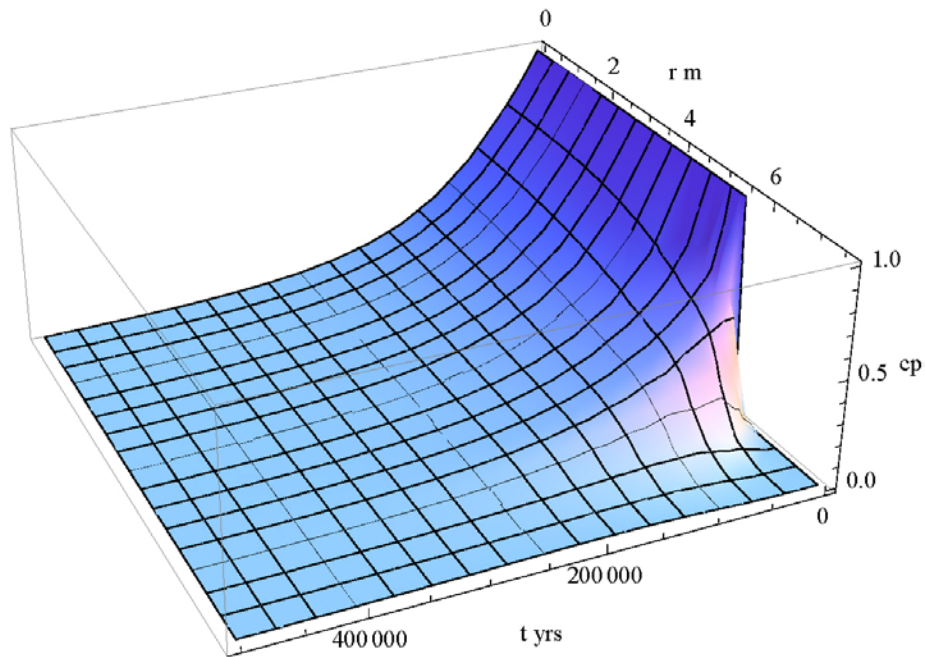


Figure 6-4. Ni-59 relative concentration c_p in waste and concrete buffer. Perpendicular flow. Results from numerical model.

Figure 6-5 shows the activity release over time for both concrete and clay buffers. The activity in the waste 1.5×10^{15} Bq in the beginning and the concentration in the pore water in the waste was $c_o = 2.3 \times 10^8$ Bq/m³ pore water. Most of the Nickel is sorbed as the retardation factor is 590 in concrete.

The migration through the buffer allows considerable decay of the nuclide during its transport from the waste to the rock. The numeric model includes the effects of the diffusion in and from the waste itself, which will influence the results. This was tested by increasing the diffusion coefficient by a factor of 5 (giving the largest possible value of the diffusion coefficient) for a system in which the waste has a radius of 5.64 m and the buffer is 2 m thick. It was found that the release rate increased by about 20%.

For the bentonite the H -parameter = 0.61. This allows very little decay during the diffusion through the bentonite. The concentration at the outside of the bentonite barrier has only dropped to $0.54 c_o$.

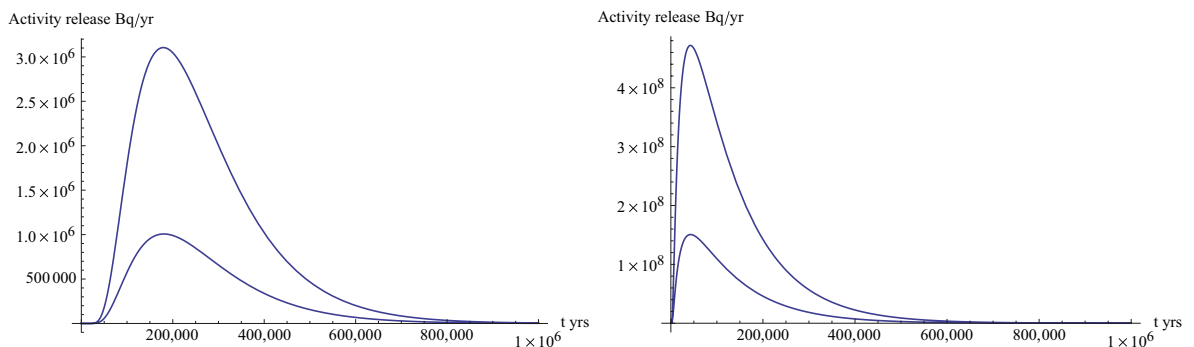


Figure 6-5. Ni-59 activity release. Concrete buffer left, bentonite buffer right figure. The lower curve is for base case hydraulic conductivity of rock. The upper is for ten times higher conductivity.

The results are sensitive to the diffusion coefficient in the buffer as can be seen in Figure 6-6. The diffusion coefficient in the degraded concrete buffer is taken to be 7.5 times larger than in the intact concrete. Also the porosity is different, 0.2 instead of 0.15. The release is about 10 times larger. This is due to the considerably smaller decay in the degraded concrete. The value of the H -parameter is 0.99 for degraded concrete, compared with 3.21 for intact concrete, which results in a higher concentration at the buffer/rock interface.

As in the previous cases the decay in the buffer determines the concentration at the buffer/rock interface and the equivalent flowrate in the rock determines the release to the seeping water.

In the following example the flow is aligned parallel to the vault. This will decrease the equivalent flowrate but only marginally influences the decay in the buffer. Figure 6-7 shows the results for this case using the same data as in the example in Figure 6-6. The release is 3 times smaller in this case. It is caused by the decrease of the equivalent flowrate, which has dropped 3 times. See row 8 in Table 5-2. The concentration at the buffer rock interface is practically unchanged and is 0.022 of the initial concentration in waste pore water.

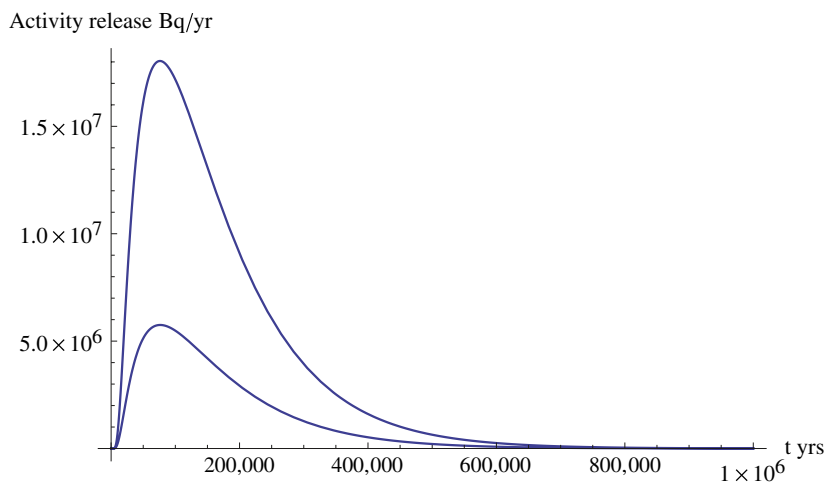


Figure 6-6. Ni-59 activity release at rock interface for a degraded concrete buffer $D_p = 5 \times 10^{-10} \text{ m}^2/\text{s}$ Perpendicular flow. The upper is for ten times higher conductivity.

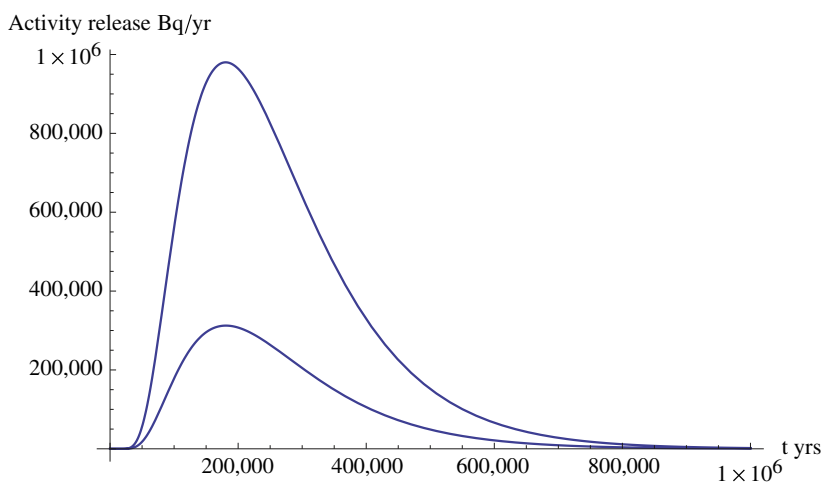


Figure 6-7. Ni-59 activity release at rock interface. The lower curve is for base case hydraulic conductivity of rock. The upper is for ten times higher conductivity. Parallel flow.

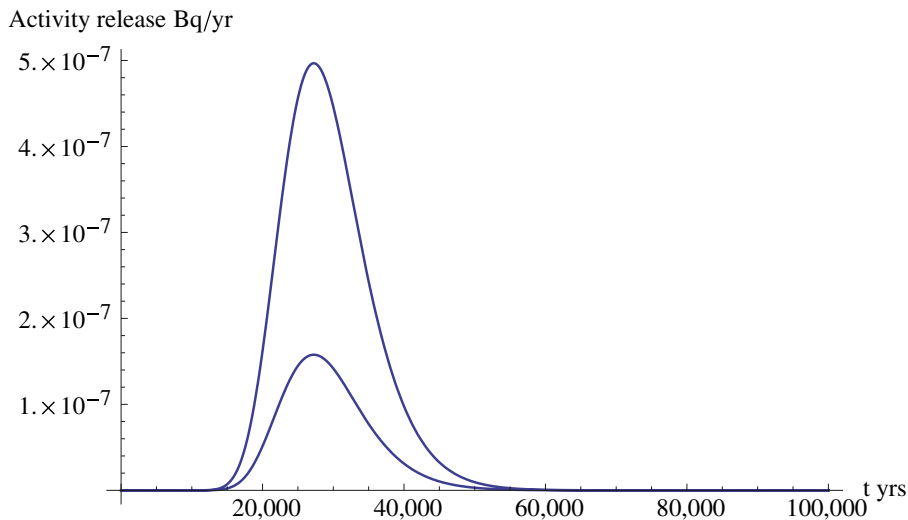


Figure 6-8. Ra-226 activity release at rock interface. The lower curve is for base case hydraulic conductivity of rock. The upper is for ten times higher conductivity. Perpendicular flow.

6.5.3 Ra-226

Ra-226 will decay considerably during its passage through the concrete buffer as can be seen by *H*-values in Table 6-1. The *H* parameter = 25, which means that the concentration at the outside of the barrier will be at most 2.1×10^{-11} of that in the pore water in the waste using Equation (6-6). A value of 1.6×10^{-11} is obtained in the numeric calculations. The release is near to vanishingly small as seen in Figure 6-8.

Main findings of Section 6.5

A model that simultaneously accounts for diffusion in the waste, buffer and the seeping water in the rock, although still very simple, is solved numerically. By accounting for the interdependence of the three barriers; waste, buffer and rock, it gives somewhat lower but more accurate results than those obtained by the simplified approach described in Section 6.3, compared with the more detailed modelling in 6.4. The examples show that when the barriers are intact, sorbing nuclides even those with fairly long half-lives, decay considerably before they reach the seeping water in the rock. The examples also show that an increase in the hydraulic conductivity of the rock by a factor of ten results in an increase of nuclide release by a factor of about 3. This is expected as the equivalent flowrate determines the carrying capacity of the nuclides and controls the escape rate and the equivalent flow rate is proportional to the square root of flow velocity (see Section 5.1). Perpendicular flow gives larger release than parallel flow, mainly because the equivalent flow rate is higher. It should be noted, however, that these models are limited to cases where there is negligible flow through buffer and waste. The impact of flow through the vault is explored in Chapter 7.

Main findings of Chapter 6

Two-metre thick concrete or bentonite buffers that have retained low hydraulic conductivities allow many of the important sorbing nuclides to decay significantly before reaching the seeping water in the rock. Concrete, even when considerably altered, retards nuclides considerably better than bentonite. For first scoping calculations the very simple formula for decay in the buffer combined with equivalent flowrate gives a very simple way to obtain first impressions of release rates. The slightly more complex model that accounts for the simultaneous effects of diffusion through waste, buffer and mobile water in the rock gives somewhat less conservative results. The results are sufficiently similar (i.e. the same order of magnitude) to support the use of the simplified model for the provisional comparison of alternative repository concepts.

7 Transport by *flow through waste* surrounded by concrete or clay

In Chapters 5 and 6 the nuclide transport by only flow or only diffusion through the concrete and clay barriers was explored in a simplified way. In the comparison of the advective and diffusive flowrates the regional hydraulic gradient was used to apply over the buffer only. There are conditions where this may not be appropriate. One such situation is when the gradient is along the vault and where the waste inside the barrier has a much larger hydraulic conductivity than the barrier. This is illustrated in Figure 7-1 below.

7.1 Simple model for flow

We develop a very simple model to estimate the flow through waste surrounded by buffer in a tunnel repository. The aim is to gain insights into how a permeable waste surrounded by a buffer with low permeability may draw in water to the repository and through the waste. The setting in Figure 7-1 is used.

A circular tunnel with radius r_b contains a cylindrical body of waste concentrically surrounded by buffer. The waste has radius r_v and extends a distance L from x_1 in the x -direction. The space between tunnel wall and waste is filled with a buffer. The hydraulic gradient is along the axis of the tunnel. The conductivities of the surrounding rock and the waste are much larger than that of the buffer. The tunnel is long compared to its radius. These conditions allow some simplifications to be made.

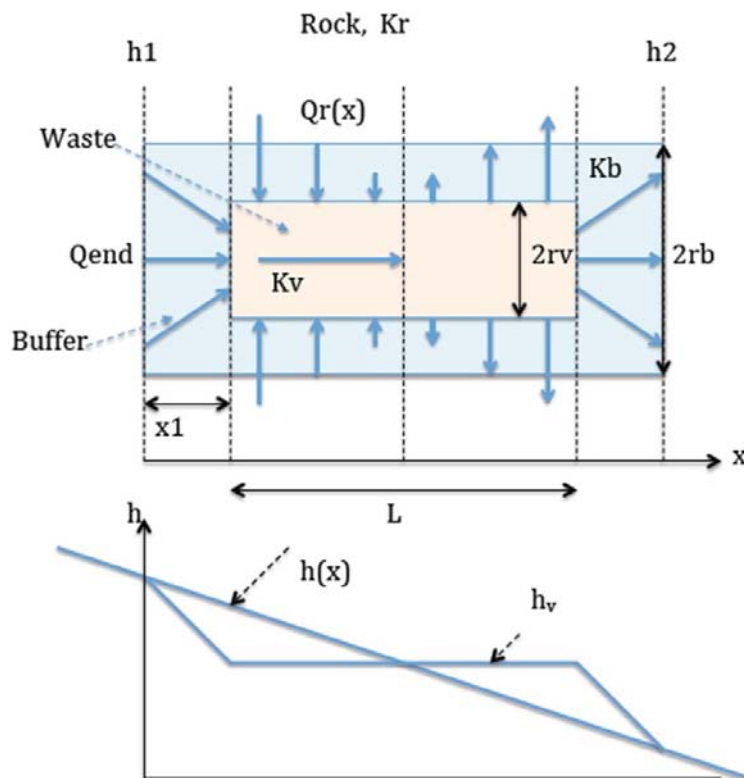


Figure 7-1. A tunnel contains waste surrounded by a buffer with low permeability.

With the hydraulic pressure h_1 in units of water head at $x = 0$ and h_2 at $x = L+2x_1$, the pressure at the rock/tunnel interface is approximately linear because of the large permeability contrast.

$$h(x) = h_1 - \frac{x}{L+2x_1}(h_1 - h_2) = h_1 - \frac{dh}{dx} x \quad (7-1)$$

In the waste, similarly because of its very high conductivity, the pressure drop for flow in the waste along the tunnel can be neglected and the pressure in the waste is constant and equal to

$$h_v = \frac{(h_1+h_2)}{2} \quad (7-2)$$

The pressure difference between rock and waste at the inlet end, x from 0 to x_1 is $h_1 - h_v$. The inflow rate, Q_{end} , of water through the mean cross section area of waste and buffer $\pi \frac{(r_b^2+r_v^2)}{2}$ at the end of the tunnel is approximated partly accounting for flow in radial direction.

$$Q_{end} = \pi \frac{(r_b^2+r_v^2)}{2} K_b \frac{(h_1-h_v)}{x_1} = \pi \frac{(r_b^2+r_v^2)}{2} K_b \frac{(h_1-h_2)}{2x_1} \quad (7-3)$$

For $x_1 < x < L+x_1$, where there is waste, the inflow is radial through the buffer, decreasing until the midpoint, where it is zero. Thereafter there is outflow as shown in the figure. The inflow rate dQ_r , radially through the buffer over a length section dx along a length of cylinder with buffer thickness r_b-r_v , and mean cross section area for flow is

$$dQ_r = -dx 2\pi \frac{(r_b+r_v)}{2} K_b \frac{(h_v-h(x))}{r_b-r_v} \quad (7-4)$$

The pressure difference $\Delta h = h_v - h(x)$ along the waste $x_1 < x < L+x_1$ is

$$\Delta h(x) = -\frac{(h_1-h_2)(L-2x+2x_1)}{2(L+2x_1)} \quad (7-5)$$

Integration from x_1 to $L/2+x_1$ gives the total inflow Q_r that comes radially into the waste up to a length section of the tunnel $L/2$. Thereafter the water flows out again from $L/2+x_1$ to $L+x_1$.

$$Q_r = K_b \frac{L^2}{8(L+2x_1)} (h_1 - h_2) \pi \frac{(r_b+r_v)}{(r_b-r_v)} \quad (7-6)$$

For $x_1 \ll L$ it reduces to

$$Q_r = K_b \frac{L}{8} (h_1 - h_2) \pi \frac{(r_b+r_v)}{(r_b-r_v)} \quad (7-7)$$

In addition there is an inflow Q_{end} to give the total flow in the tunnel of

$$Q_{tot} = \pi \frac{(r_b^2+r_v^2)}{2} K_b \frac{(h_1-h_2)}{2x_1} + K_b \frac{L^2}{8(L+2x_1)} (h_1 - h_2) \pi \frac{(r_b+r_v)}{(r_b-r_v)} \quad (7-8)$$

The pressure difference $h_1 - h_2$ is determined by length of the tunnel and the hydraulic gradient, which is assumed to be known. Equation (7-1), slightly rewritten gives

$$h_1 - h_2 = \frac{dh}{dx} (L + 2x_1) \quad (7-9)$$

Introducing the gradient instead of the pressure difference gives

$$Q_{tot} = \pi \frac{(r_b^2+r_v^2)}{2} K_b \frac{dh}{dx} \frac{(L+2x_1)}{2x_1} + K_b \frac{dh}{dx} \frac{L^2}{8} \pi \frac{(r_b+r_v)}{(r_b-r_v)} \quad (7-10)$$

It may be noted that the flowrate through the waste is determined by the conductivity of the buffer and is independent of that of rock and waste given the assumptions underlying the model. It may also be noted that the contribution to flowrate from the second term increases proportionally to the tunnel length squared, and this is the dominating term for a long tunnel.

For flow perpendicular to the tunnel the situation is quite different. The low permeability barrier surrounds the permeable waste. This diverts the flow in the rock around the cylindrical tunnel. The flowrate through the tunnel can be approximated by

$$Q_{perp} \cong L2r_b K_{av} \frac{dh}{dx} \quad (7-11)$$

where

$$K_{av} = \frac{d_b + r_v}{\frac{d_b}{K_b} + \frac{r_v}{K_v}} \quad (7-12)$$

$d_b = r_h - r_v$ is the thickness of the buffer. A large fraction of this flowrate will also pass the waste if $K_v \gg K_b$. In this case the flowrate increases proportionally to the tunnel length and not to the square of the length as when the gradient is along the tunnel.

Equations (7-11) and (7-12) have been compared with the exact more complex solution supplied by Paul Smith (2013, personal communication). The above simple equation underestimates the flow through the tunnel by less than a factor of two for the cases of interest, i.e. when the waste has a high conductivity compared to the buffer and when the buffer has low conductivity compared to that of the surrounding rock.

In this case the flowrate increases proportionally to the tunnel length and not to the square of the length as when the gradient is along the tunnel.

When the tunnel with the highly conductive waste is aligned at an angle α ($\alpha = 0$ for gradient perpendicular to the tunnel) to the hydraulic gradient the flowrate through the waste can be approximately estimated by

$$Q_{tot,\alpha} = Q_{perp} \cos(\alpha) + Q_{tot} \sin(\alpha) \quad (7-13)$$

As the flowrate is much larger for the parallel than for the perpendicular case this implies that the higher flowrate Q_{tot} for parallel flow is a better choice if one wants to be conservative. A 45 degree angle would decrease the flowrate only by 30%, a 30 degree angle by 50%.

Main findings of Section 7.1

A highly conductive waste embedded in a low conductivity buffer will draw in water through the buffer if the gradient is more or less parallel with the vault. Considerably larger flows can result than if waste and buffer have similar conductivities. The simple formulas give the flowrate through a very permeable waste when it is embedded in a low conductivity buffer at all intersection angles of vault and hydraulic gradient. Both flow through the plugs at the ends of the vault and radially through the buffer are obtained. If the waste has a higher permeability than the buffer, a considerably higher flowrate through the waste can be expected the more the gradient is parallel to the vault.

7.2 Residence time and decay in the buffer

A nuclide decays exponentially with time. The residence time in the buffer for a sorbing nuclide is $t = t_w R_a$, where t_w is the water residence time in the buffer. For the plug at the end of the tunnel the flowrate is given by Equation (7-3). Using the same mean cross section as for the flow, $\pi \frac{(r_b^2 + r_v^2)}{2}$ the volume of the water in the plug is approximated by

$$V_{w,end} = x_1 \pi \frac{(r_b^2 + r_v^2)}{2} \varepsilon_b \quad (7-14)$$

(7-14) with (7-3) gives

$$t_{w,end} = \frac{2x_1^2 \varepsilon_b}{(h_1 - h_2)K_b} \quad (7-15)$$

For solubility limited conditions and if there were no diffusion, and assuming that the concentration c_o in the waste is not depleted much by the out-transport due to the flow, the concentration in the water leaving the plug is

$$c_{end} = c_o e^{-\lambda R_a t_{w,end}} \quad (7-16)$$

This applies if the simultaneous transport by molecular diffusion is small in comparison, i.e. when the flow is characterised by plug-flow. This will be explored below. Similarly the residence time for the water flowing radially through the buffer is

$$t_{w,r} = \frac{(r_b - r_v)^2 \varepsilon_b}{(h_v - h(x))K_b} \quad (7-17)$$

As the flowrate varies with location along the tunnel the residence time also depends on the location, Equation (7-4). There will be less decay near the end of the tunnel. The mean concentration in the water that flows radially out of the tunnel Q_r , Equation (7-6), is

$$c_{r,mean} = \frac{c_o}{Q_r} \int_{x_1+L/2}^{x_1+L} \frac{dQ_r(x)}{dx} e^{-\lambda R_a t_{w,r}(x)} dx \quad (7-18)$$

Equation (7-18) has a complicated analytic solution but can readily be integrated numerically. The mean concentration in the water through the end plug and by the radial flow is

$$c_{mean,tot} = \frac{Q_{end} c_{end} + Q_r c_{r,mean}}{Q_{end} + Q_r} \quad (7-19)$$

7.2.1 Simultaneous flow and diffusion

The above relations show the carrying capacity of the flowing water if there were no simultaneous diffusion or when nuclide transport by flow dominates over that by diffusion.

A simple estimate of the relative rates of transport by flow and molecular diffusion can be made by comparing the characteristic times for flow and diffusion. The characteristic time for flow for a nuclide with retardation factor R_a is

$$t_{c,flow} = R_a t_w = \frac{R_a d}{u} \quad (7-20)$$

d is the distance travelled by the water with velocity u . Equation (7-20) gives the time of nuclide arrival. If this occurs by plug-flow the concentration rises suddenly from 0 to the value given by Equation (7-15). When there also is diffusion the front is not sharp but rises gradually already before the characteristic time for flow. The characteristic time for diffusion is

$$t_{c,diff} = \frac{d^2}{D_a} = \frac{d^2 R_a}{D_p} \quad (7-21)$$

The ratio of diffusion-time to flow-time, Pe , is called Peclet number

$$Pe = \frac{ud}{D_p} \quad (7-22)$$

When $Pe > 100$ flow dominates the transport. When $Pe < 1$ diffusion dominates.

At low Peclet numbers, that is when transport is *dominated by diffusion*, the concentration at the outside of the buffer is not much influenced by the flow. However, the flow through the waste and buffer may contribute much to the release because the flowrate will carry nuclides with a concentration equal to that at the outside of the buffer. The latter is determined mostly by diffusion. Then the

escape by diffusion and by flow is additive by Equations (5-1a) and (5-1c) and the concentration in both equations is the same $c_{exit} = c_{RI}$. As c_{water} for most nuclides is zero the release is

$$N_{tot} = (Q + Q_{eq})c_{RI} \quad (7-23)$$

If the flow through the buffer does not influence the concentration at $x = x_o$ (the buffer rock interface) then Equation (7-23) is a fair approximation. This is explored in the examples below.

Main findings of Section 7.2

For flow parallel to the vault simple formulas can be used to assess the impact for flow through the end of the vault as well as for radial flow through the buffer at all locations along the vault. Another simple formula gives the flow through the waste when flow is perpendicular to the vault. When water flows through the buffer and waste the nuclides carried with it will be retarded during the passage of the water through the buffer and given time to decay. A very simple criterion is presented that shows when nuclides *carried by flow* through the buffer may add to the transport by the simultaneous diffusion that always is present. See text above Equation (7-23)

7.3 Model for transport by simultaneous flow and diffusion

In order to assess when the flow through the buffer can become important the following simple model is used. It describes how the nuclide is simultaneously transported by *flow and diffusion* through the buffer. The concentration c_{exit} at the outlet end times the flowrate Q gives the rate at which the nuclide is carried away by the water that passes through the buffer. It is described by Equation (5-1a). This can be compared to the rate of nuclide transport by the equivalent flowrate Q_{eq} times the concentration c_{RI} at the outside of the buffer at the rock interface where there is no outflow of water. It is expected that at low Peclet numbers when diffusion dominates the transport through the buffer, c_{exit} will be only slightly increased above c_{RI} . This would show that Equation (7-23) can be used for low Peclet numbers, which simplifies the computations considerably. The following equations are solved for the flow case

$$R_a \frac{\partial c_p}{\partial t} = -u \frac{\partial c_p}{\partial x} + D_p \frac{\partial^2 c_p}{\partial x^2} - \lambda R_a c_p \quad (7-24)$$

The initial condition is

$$c_p = 0 \text{ for } t = 0 \text{ and } 0 < x < x_o \quad (7-25)$$

The boundary conditions are

$$c_p = c_o \text{ for } x = 0 \text{ for } t > 0 \quad (7-26)$$

and

$$\frac{\partial c_p}{\partial x} = 0 \text{ at } x = x_o \text{ for } t > 0 \quad (7-27)$$

x_o is the thickness of the barrier. It can be the thickness of the buffer surrounding the waste around the vault or the thickness of the buffer at the end of the vault. The equations are solved numerically although there exist an analytical solution (Kumar et al. 2009), which is lengthy, however. The nuclide outflow *by flow and diffusion* then is obtained from Equation (5-1a) with $c_{exit} = c_p$ at $x = x_o$. In the above case the presence of the flow in the buffer influences the results and the concentration in the buffer increases more rapidly than when there is no flow. In the Q_{eq} case, where there is no flow through the buffer, the nuclide only diffuses through it to x_o from which location it is carried away by the equivalent flowrate Q_{eq} . The equation with initial and boundary conditions are

$$R_a \frac{\partial c_p}{\partial t} = D_p \frac{\partial^2 c_p}{\partial x^2} - \lambda R_a c_p \quad (7-28)$$

$$c_p = 0 \text{ for } t = 0 \text{ and } 0 < x < x_o \quad (7-29)$$

$$c_p = c_o \text{ for } x = 0 \text{ for } t > 0 \quad (7-30)$$

$$D_e A \frac{\partial c_p}{\partial x} = -Q_{eq} c_p \text{ at } x = x_o \text{ for } t > 0 \quad (7-31)$$

A is the outer surface area of the buffer.

7.4 Some examples

7.4.1 Flowrates

Base case data are used in the examples. Figure 7-2 below shows the flow through the waste in a tunnel aligned in the direction of the gradient as a function of the *length of tunnel*, (not the coordinate along the tunnel). The buffer consists of clay. The rapidly disappearing curve in the lower left hand corner is for waste with the same conductivity as the buffer. The straight line near the x-axis is the flowrate that enters horizontally through the 2 m thick clay at the inlet end of the tunnel. The bent curve shows the flowrate that enters radially into the waste. For a 200 m long tunnel the total flowrate that enters (and then leaves) the tunnel is 1.06 m³/yr. This compares reasonably well with results obtained using a high-resolution numerical solution, which gave $Q_{tot} = 0.51 \text{ m}^3/\text{yr}$. In that calculation the conductivities of waste, bentonite and rock were 10^{-6} , 10^{-9} and $3 \times 10^{-9} \text{ m/s}$ respectively. The contrast in conductivity between buffer and rock is only 3, which is not considered to be very large and thus does not quite fulfil the assumption in the simple model. The simple model will, as was found, overestimate the flowrate in this case. For concrete with a much lower hydraulic conductivity the flowrates by the simple model and more elaborate numerical calculations were much closer.

Figure 7-3 shows the results for a concrete buffer. The flowrate is much smaller due to the lower conductivity of concrete. The total flowrate with the simple model for a 200 m long tunnel is 0.053 m³/yr. Numerical simulations gave 0.052 m³/yr. In this case the assumptions for the simple model are better fulfilled than for the bentonite case. When the gradient is perpendicular to the tunnel the total flowrate is 0.011 m³/yr, (Numerical model 0.01 m³/yr).

Table 7-1 summarises the results. A numerical finite element code was used to solve the Darcy equation for flow through the rock, buffer and waste for comparison. As expected the results agree better for the concrete case because the contrast between the hydraulic conductivity and the rock is larger than for the clay case. In the simple model it is assumed that the hydraulic conductivity of the buffer is much smaller than that of the rock.

Q in waste at $x=L/2 \text{ m}^3/\text{yr}$

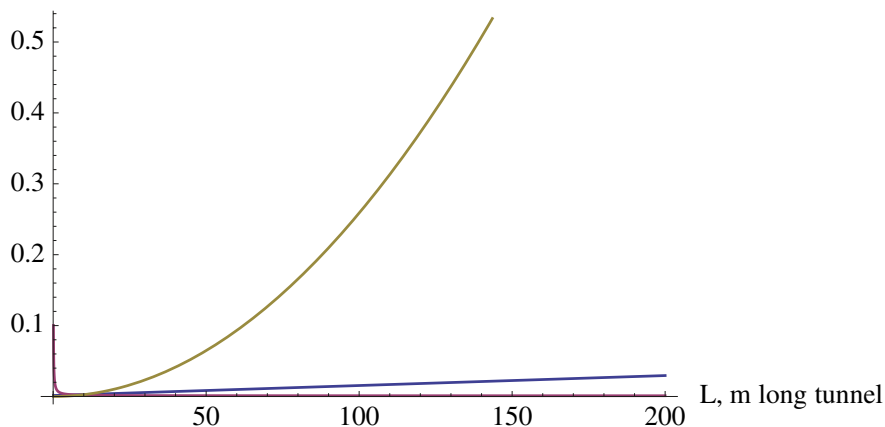


Figure 7-2. Flowrate through waste in different length tunnels with bentonite buffer.

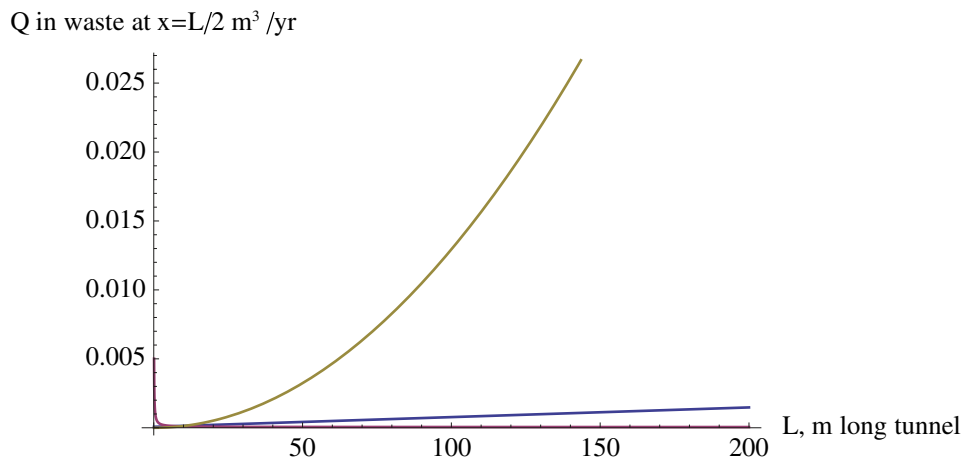


Figure 7-3. Flowrate through waste in different length tunnels with concrete buffer.

Table 7-1 Summary of flowrates through waste, m³/yr.

	Parallel simple	Parallel numerical	Perpendicular simple	Perpendicular numerical
Concrete	0.052	0.052	0.011	0.010
Bentonite	1.05	0.51	0.12	0.015

7.4.2 Nuclide escape by flow

Figure 7-4 shows the rate of escape of Ni-59 for the base case conditions when flow is perpendicular to the vault. The concentration c_0 is taken to be 1 mol/m³ porewater at the inlet to the buffer. The Peclet number for this case is 0.011. In this case $Q = 0.005$ m³/y and $Q_{eq} = 0.075$ m³/yr.

As shown in Figure 7-5 the relative porewater concentration c_{exit} is practically indistinguishable from c_{Ri} . The relative pore water concentration is the ratio between the concentrations on the outside of the buffer to that on the inside.

Figure 7-6 shows the release for a 20 times higher flowrate through the concrete buffer, which would result from a concrete with 20 times higher hydraulic conductivity in the buffer. The sorption coefficient is assumed to be the same.

The Peclet number in the high flowrate case is 0.23. Figure 7-7 shows the porewater concentration for the two cases. c_{exit} , the upper curve, is somewhat larger than the concentration that results from only diffusion. The example was based on Ni-59. Similar small differences are obtained for Cl-36 and Ra-226.

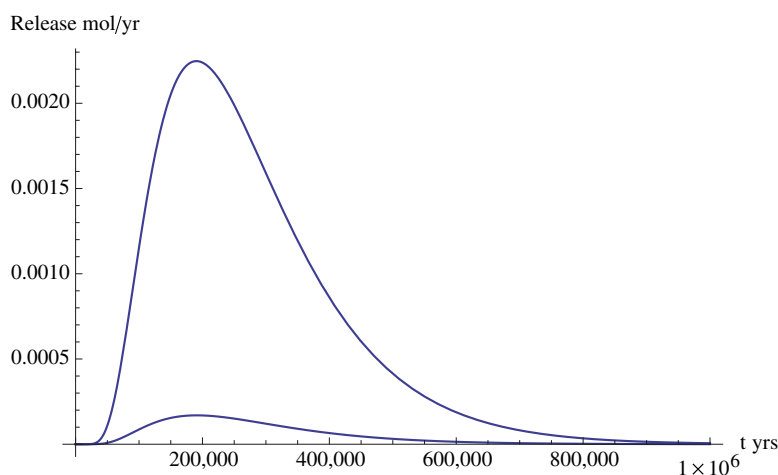


Figure 7-4. Release by flow, lower curve and by Q_{eq} , upper curve.

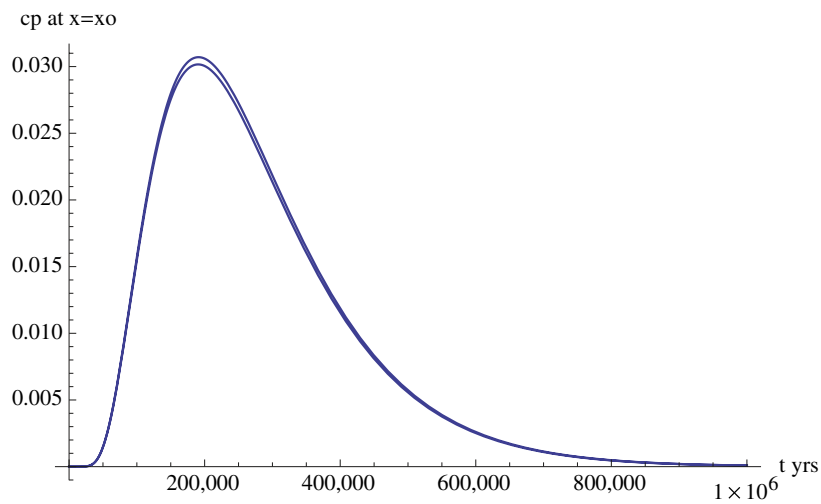


Figure 7-5. Pore water relative concentration c_p at $x = x_0$. Upper curve for flow case and lower curve for Q_{eq} case.

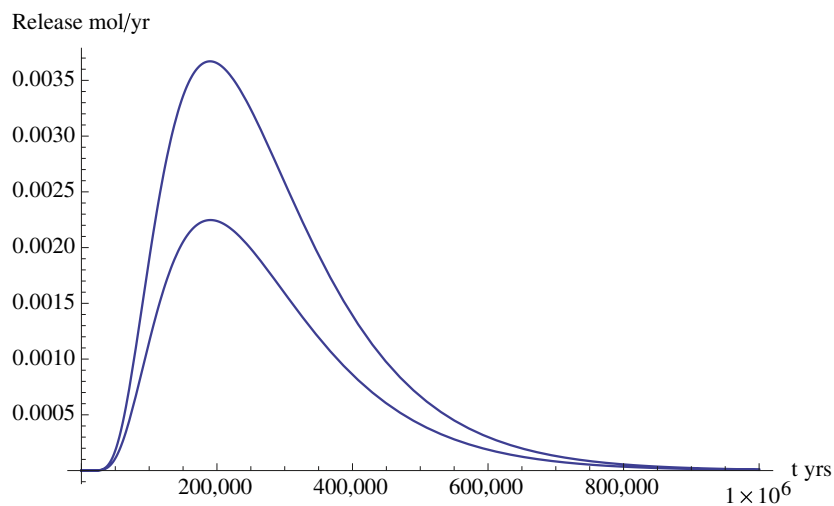


Figure 7-6. Release by flow, upper curve and by Q_{eq} lower curve. High flowrate.

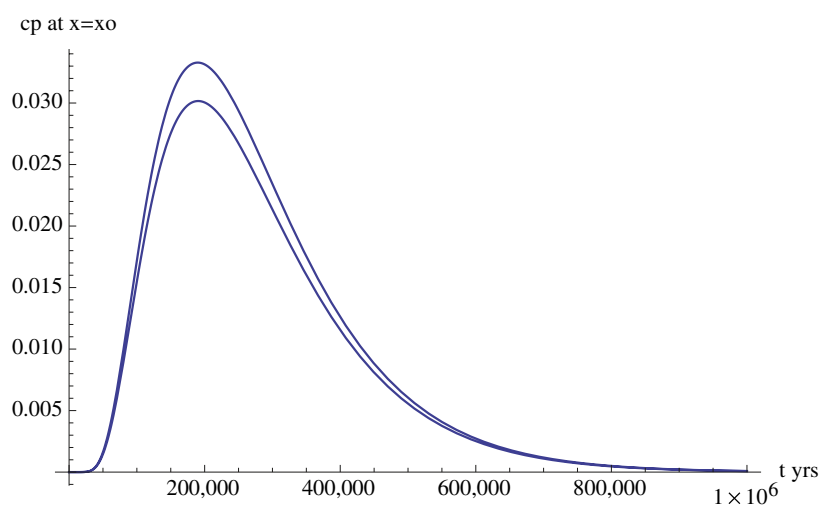


Figure 7-7. Pore water concentration at $x = x_0$. Upper curve for flow case and lower curve for Q_{eq} case. High flowrate.

7.4.3 Flowrates, residence times and Peclet numbers

Table 7-2 summarises the flowrates, the water residence times and the Peclet numbers for the tunnel and Supersilo designs. In all cases the Peclet numbers are well below 1. In all cases release by diffusion through the buffers will dominate the nuclide release through the buffer. However, for flow parallel to the tunnel and with bentonite the flowrate through the waste can considerably increase the release to the rock. This is seen by the large radial flow 1.03 m³/yr in Table 7-2. This flow will carry a nuclide concentration equal to that in Q_{eq} as the transport through the buffer is dominated by diffusion. The table shows equivalent flowrates Q_{eq} as well as flowrates through the waste Q . In addition water residence times for flowing water through the buffer and Peclet numbers are presented. For parallel flow the flowrates through the end part of the buffer as well as the flowrate that passes radially through the buffer are given.

Main findings of Section 7.4

For parallel as well as for perpendicular flow to the vault simple formulas can be used to assess the impact of the presence of a very permeable waste. This also applies to the Supersilo. The concentration at the rock interface is essentially determined by diffusion through the buffer, even in the presence of flow through the waste and buffer. It is also shown that the additional simplification for the decay during the diffusion through the buffer using the model in Section 6.1, $c_{xo} = c_o e^{-H}$, overestimates the release compared to the results using the more elaborate model in Section 6-3. It may be used to compare different design alternatives because the transport through the buffer(s) is only marginally influenced by flow, which can be quite different in the different cases.

Main findings of Chapter 7

Even in the case when there is considerable flow through the waste many sorbing nuclides will be retarded and decay to insignificance during their passage through the buffer. Although flowrates through the waste and buffer can be on the order of m³/yr to carry nuclides out through the buffer the transport by molecular diffusion is more rapid than by flow and will largely determine the overall release rate.

However, should there be one or more highly transmissive fractures *in the buffer* through which the water flows out the nuclides would not be retarded by sorption and would escape with the flowing water without much decay. This could lead to a considerable increase in nuclide release. Concrete is probably more vulnerable than bentonite in this respect.

Table 7-2. Flowrates, water residence times and Peclet numbers for the different design options.

	Bentonite	Concrete
Perpendicular flow to tunnel		
Qeq m ³ /yr	0.075	0.075
Q m ³ /yr	0.11	0.0055
tw yrs	22,100	166,000
Pec	0.023	0.011
Parallel flow with tunnel		
Qeq m ³ /yr	0.023	0.023
QRadial m ³ /yr	1.03	0.052
QEnd m ³ /yr	0.03	0.0015
QSum	1.06	0.0535
twRadial yr	3,200	24,200
twEnd yr	9,600	72,000
PecRadMax	0.32	0.16
PecEnd	0.32	0.16
Supersilo		
Qeq m ³ /yr	0.041	0.041
Q m ³ /yr	0.093	0.0046
tw yrs	8,800	65,600
Pec	0.058	0.029

8 Degradation of concrete and bentonite

Concrete contains sodium, potassium and calcium hydroxide. These generate very high pH in the concrete porewater. When concrete is in contact with bentonite, these compounds dissolve and diffuse out of the concrete and into the bentonite. The smectite in the bentonite reacts with the hydroxide and forms non-swelling minerals. The large amounts of calcium in the concrete can also transform some of the sodium smectite to calcium smectite, which is more permeable and swells less. Both concrete and bentonite may lose their desirable buffer properties.

Many experimental and modelling studies have been made to investigate the rate of degradation of concrete and bentonite. The results of some recent studies are described in Neretnieks (2013, manuscript to be published). Degradation distances ranging from tens of centimetres to meters have been predicted over a 100,000 year period. This wide span of predicted distances is due to the use of different assumptions on the diffusion properties of the media, on reaction rates and on the choice of secondary minerals that are allowed to form. The choice of minerals is often quite subjective.

A very simple model that can be used to simulate the mutual degradation of the media was developed and is presented in Neretnieks (2013, manuscript to be published). It describes the rapid, solubility limited, dissolution of calcium hydroxide and its rate of diffusion out of the concrete to the interface to the bentonite. From this interface the calcium hydroxide diffuses into the bentonite and reacts rapidly with the smectite, which turns into secondary non-swelling minerals. A dissolution front develops and recedes in the concrete and a reaction front advances outward in the bentonite. The rate is determined by the diffusivity in the media, the abundance of hydroxide and smectite in the media. Figure 8-1 illustrates the process.

All reactions are assumed to be very rapid compared with the time scales the model will be used for. It is therefore deemed to be conservative in this respect. The model results compare fairly well with those obtained using much more complex models including tens of minerals as well as reaction kinetics. Using the base case data the rate of simultaneous degradation of concrete and bentonite is shown in Figure 8-2. This is based on the diffusion properties of unaltered media.

The diffusion takes place in the *altered zone* of the media. It is probable that the diffusion properties in the altered zone are different than in the unaltered media. Should the diffusivity have increased by a factor 10 in the degraded regions, the fronts will move faster. This is shown in Figure 8-3.

A low pH cement decreases the rate of front propagation because it contains much less portlandite. This degradation case is shown in Figure 8-4.

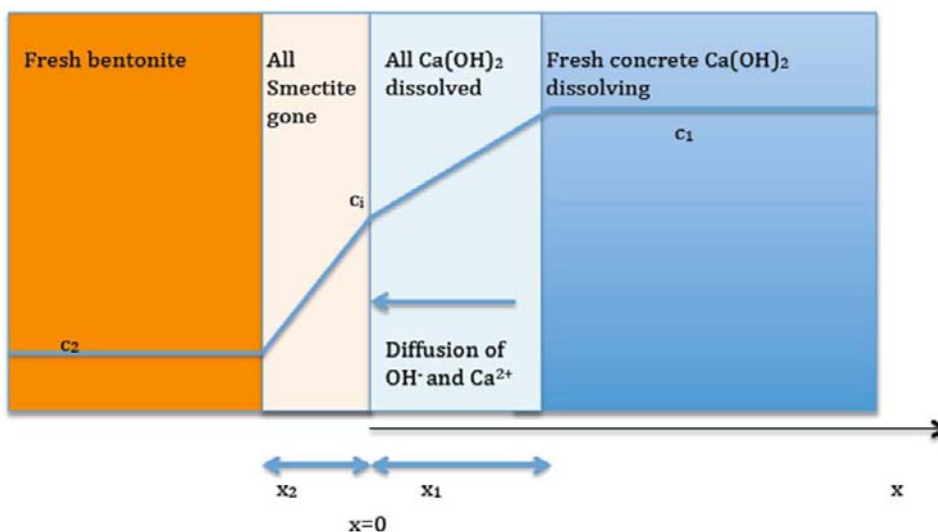


Figure 8-1. Depletion of Portlandite in concrete and smectite in bentonite.

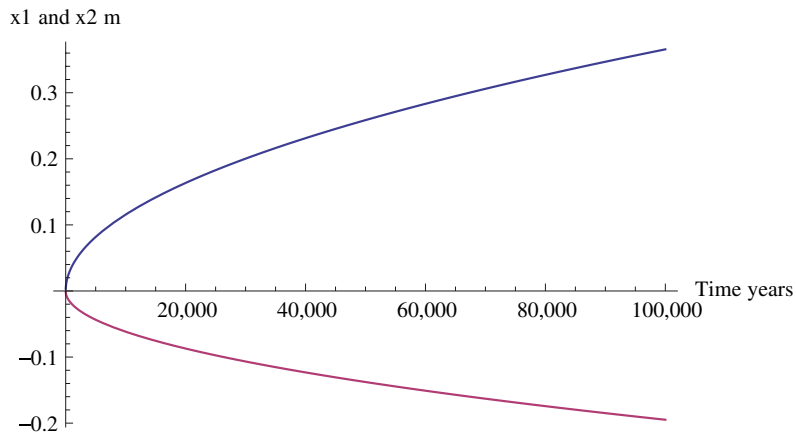


Figure 8-2. Evolution of the zones reaction fronts over time. Diffusion coefficients of unaltered media. Upper curve for concrete.

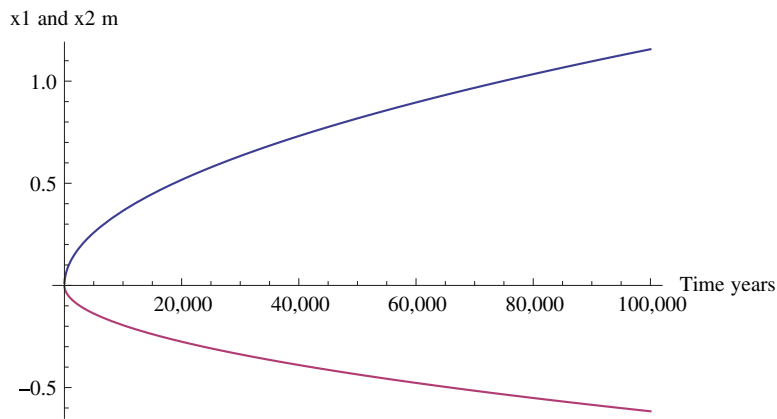


Figure 8-3. Evolution of the zones over time. Diffusion coefficients of altered media 10 times larger than the base case. Upper curve for concrete.

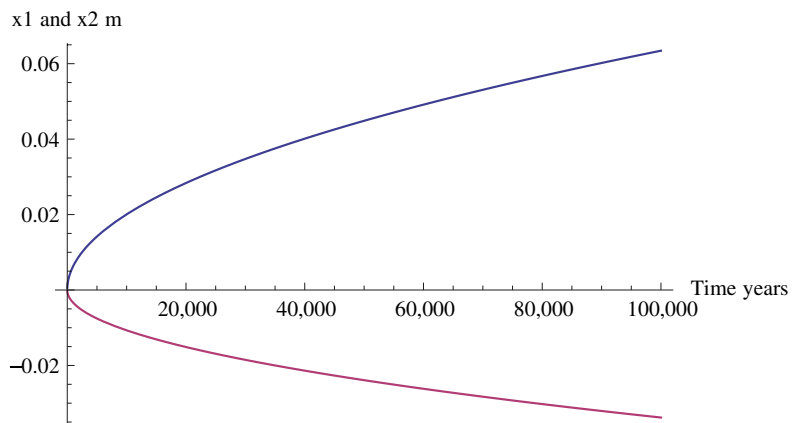


Figure 8-4. Evolution of the zones over time. Base case but low pH cement. Upper curve for concrete.

There are several simplifying assumptions underlying this simple model. However, it gives results that are in the range of results obtained by other much more complex and elaborate models. The central mechanisms that drive the whole degradation process, namely hydroxyl migration and reactions, are the same in the simple model and in the other models. The simple model should therefore be useful for scoping calculations when one wants to gain an impression of how a change in hydroxide or smectite content, or diffusion properties influence the development of the degradation rates. The results of the simple simulation suggest that low pH cements are preferable for use in the waste encapsulation as this decreases the rate of degradation of bentonite. Similarly, should a Supersilo be considered, this also may be made of low pH cement. The formation of ettringite in the concrete by intruding sulphate can on the one hand clog pores and stop further transport and therefore degradation. On the other hand the swelling ettringite can fracture the concrete from within and counteract the clogging. This has not been modelled.

Main findings of Chapter 8

Concrete and bentonite in contact with each other will lead to a slow degradation of both media. The use of a conventional Portland cement, which contains large amounts of hydroxide can lead to degradation up to meter(s) in the concrete stabilised waste as well as in the bentonite used as buffer over a 100,000 year period. Using low pH cement can limit the damage to some tens of centimetres. The same applies for the Supersilo.

9 Comparison of the design options

9.1 Tools used

The bentonite and concrete buffers have two important functions, namely to act as barriers to *flow* and to *diffusion* of radionuclides escaping from the waste. To compare the escape rates from the different barriers a set of models have been devised. The models have been made simple in order to highlight those mechanisms that have the largest impact on the release rates. Although diffusion and flow interact it was found that it is sufficient to assess the escape by flow and that by diffusion separately and add the two escape rates. This facilitates the comparison of the design options considerably. This approach also points to specific weaknesses in each design option.

A first impression of how the water flowrate through the vault depends on the hydraulic properties of the barriers can be gained from the data in Table 5-2. Firstly in lines 1 to 5 it is seen from the data in the two columns that the water flowrate through a vault depends on at what angle the vault is in relation to the hydraulic gradient. It is also seen that the presence of a prominent fracture and a fracture zone could increase the water flowrate through the vault considerably if the buffer has been much degraded; lines 6 and 7. The flowrate through the buffer will carry the nuclides with it in a concentration that depends on how they were released from the waste and how they decayed during their transport through the buffer. It was found by the modelling that in all cases molecular diffusion is the main transport mechanisms for nuclides migrating from the waste through the buffer and will determine the concentration in the flowing water leaving the buffer. This simplifies modelling considerably as the rate of nuclide transport by diffusion also can be assessed by a simple approach.

In addition to the transport by the water that flows *through the waste and buffer*, Q , also the water that flows past the buffer, or in the case of the hydraulic cage, past the waste, takes up nuclides by molecular diffusion in the water *flowing past the buffer or waste*. The transport capacity by this mechanism is determined by the equivalent flowrate Q_{eq} . The value of this entity can also be obtained by a very simple expression. The flowrate Q and Q_{eq} will both carry the nuclide in the same concentration, c_{RI} , and the total nuclide flowrate is the sum of these. An upper bound of the concentration at the buffer-rock interface c_{RI} can be estimated by another simple expression that accounts for diffusion, retardation and decay in the buffer.

Table 9-1 summarises the expressions used to obtain the entities described above.

Table 9-1 Summary the expressions used to obtain flowrates and concentrations.

	Section	Equation(s)	Comments
Flowrate Q through vault w. same Kh in waste and buffer, all directions	5.2.4	In Holmén (1997)	Used to obtain an impression of impact on flowrates through vault of different contrasts in hydraulic conductivity and approach angles.
Flowrate Q through vault w. different Kh in waste and buffer	7.1		A very important quantity that influences the release of nuclides from the vaults if barriers should attain high hydraulic conductivity by fracturing or degradation.
Perpendicular flow		7-11	Through buffer-waste-buffer.
Parallel flow		7-3 and 7-6	Through end of tunnel and radially through buffer.
Q_{eq} for flow in rock past buffer	5.2.3		A very central quantity that together with Q determines the carrying capacity of nuclides from the vaults. It only depends on rock properties.
Perpendicular flow		5-6	For the Supersilo both Equation 5-6 and 5-7 are used.
Parallel flow		5-7	
Q_{eq} for flow in Hydraulic cage	5.3	5-10	It is found that all water flowing through the hydraulic cage becomes fully contaminated.
Concentration c_{RI} at buffer-rock interface			This is a central entity as it determines the nuclide concentration in both Q and Q_{eq} . It only depends on the properties of the buffer and the nuclide.
Buffer	6.1	6-6	This gives an upper bound of the maximum concentration of c_{RI} at the outside of the buffer. It will decrease with time as the nuclide is depleted in the waste and as it decays.
Hydraulic cage	5.3.1	5-8 and 5-9	The concentration in the waste is the driving force for uptake in the flowing water. No resistance in the waste is accounted for.

9.2 Comparisons

First the pros and cons of the different design options are discussed. Thereafter comparisons are made between them.

9.2.1 Hydraulic cage

In the hydraulic cage all the water that flows in the cage will take up the nuclides at the rate that the waste can release it. The flowrate in the hydraulic cage is entirely governed by hydraulic conductivity of the rock including that of prominent fractures and any fracture zone intersecting the vault. The hydraulic cage is not a barrier; on the contrary, it promotes the release of radionuclides. The highest water flowrates in row 1 and 2 (in Table 5-2) plus the flowrates in a prominent fracture and zone will carry the nuclides. This can be on the order of tens of m^3/yr .

9.2.2 Tunnel with concrete buffer

The flowrate through the vault with a non-degraded and unfractured concrete buffer permits very little flow, $< 0.01 \text{ m}^3/\text{yr}$, through the vault provided also the waste has the same low hydraulic conductivity. The release will be governed by the equivalent flowrate. This is on the order of less than $0.1 \text{ m}^3/\text{yr}$ (Table 5-1 or row 8 in Table 5-2). In addition the equivalent flowrates in any prominent fracture and fracture zone need to be added. This may result in a total carrying capacity of less than $0.2 \text{ m}^3/\text{yr}$.

If the waste has a much higher hydraulic conductivity than the good concrete the flowrate *through the buffer and waste* can increase considerably if the hydraulic gradient is aligned parallel to the vault. The flowrate increases to more than $0.05 \text{ m}^3/\text{yr}$ (Table 7-2). The total carrying capacity is less than $0.25 \text{ m}^3/\text{yr}$ but that by Q_{eq} still dominates. A possible concern is that if the concrete develops some highly transmissive fractures, and when the waste is very permeable, considerably higher flowrates can develop. This is especially important if the concrete fractures are adjacent to a prominent fracture in the rock or a fracture zone. A total flowrate on the order m^3/yr through the waste can result (Table 5-2, rows 6 and 7). In such fractures in the concrete also the retardation of the nuclide is not effective. The retardation can become negligible when the water flows rapidly through a fracture in the buffer.

When the concrete is intact molecular diffusion dominates the transport. Concrete has excellent retardation properties for all sorbing nuclides. Many of the strongly sorbing nuclides will decay to insignificance in the concrete. This can be seen in Table 6-1, where several H -values in columns 4 and 5 are larger than 20. For an H -value of 20 the nuclide has decayed by 9 orders of magnitude at the outside of the buffer compared to that on the inside. Even a concrete that has degraded and in which the pore diffusion coefficient has increased tenfold (column 5) the retardation properties are better than in bentonite.

9.2.3 Tunnel with bentonite buffer

The hydraulic conductivity of bentonite in the base case is 20 times higher than in good concrete. This implies that also the flowrate through the vault will increase by nearly this amount. This is especially prominent when the tunnel is aligned parallel to the hydraulic gradient and the waste has a much higher hydraulic conductivity than the bentonite. The flowrate can then increase to more than $1 \text{ m}^3/\text{yr}$ (Table 7-2). The equivalent flowrate is the same as for concrete i.e. about $0.02 \text{ m}^3/\text{yr}$. The nuclide transport rate will be dominated by flow.

The large increase in flowrate in the parallel flow case can be decreased if the vault is divided in two or more vaults. For parallel flow the flowrate through a vault with a very permeable waste increases as the square of the length. Dividing the vault in two will decrease the flowrate in each half by a factor of 4. This would decrease the sum of flowrates in the two vaults by a factor of 2.

The retardation in bentonite is not as strong as in concrete. Nevertheless, many actinides and tri-valent fission products have large H -values. The H -value is proportional to the thickness of the buffer. In the base case it was taken to be 2 m. Increasing the thickness of the buffer can increase the decay.

The flow and retardation properties of bentonite are not as favourable as those of unaltered concrete. However, bentonite is deemed to be less prone to develop fractures than concrete. On the other hand bentonite will be in direct contact with concrete in the waste and waste containers. Concrete and bentonite in contact degrade each other over time. This will decrease the retention properties of both materials. Although no retention properties in the waste has been accounted for, in practice it may impede release to some degree.

Bentonite can be lost by chemical erosion if exposed to larger flowrates of fresh water during long times. This is not the case for concrete although leaching of its alkali and earth-alkali hydroxides may alter it.

9.2.4 Supersilo having either concrete or bentonite as buffer

The dimensions of the Supersilo, if made as a cylinder with the same diameter as height is 33.4 m. This includes a 2 m thick buffer and a 2 m thick concrete wall surrounding the waste on all sides. The flowrate through such a vault is essentially independent of the flow direction. The flowrate through the buffer and waste is 0.008 and 0.16 m³/yr for concrete and bentonite respectively. The equivalent flowrate is 0.11 m³/yr. The decay during passage through the buffer is slightly larger as that in tunnel with 2 m concrete because of the presence of the bentonite. It may be easier to avoid an intersection of a fracture zone using the silo alternative. A Supersilo has both bentonite and concrete buffers. Either of them can be closest to the waste. The flowrate through the silo will be most influenced by the buffer with the lowest hydraulic conductivity. With 2 m thickness of both barriers the flowrate is less than 0.01 m³/yr. The decay in the buffers is also governed by the buffer with the largest *H*-value. One may even, as an approximation, add the *H*-values of both barriers. If the bentonite barrier is nearest the rock any fractures developed in the concrete will not be able to carry much water because the bentonite limits the flowrate in to the concrete. The retardation in the concrete will thus also not be influenced by fracturing. Both flow and retardation in the concrete will therefore be favourably influenced by a silo construction with both barriers. However the direct contact between concrete and bentonite will promote degradation of both buffers.

Placing the bentonite between waste and concrete will lead to attack on the bentonite from both sides. Fractures that develop in the outer concrete silo wall, if more transmissive than in the rock, will allow water to flow to the bentonite and thus short-circuit the diffusion barrier in the concrete.

9.3 Degradation of concrete and bentonite

The degradation of concrete and bentonite by fresh or saline water flowing past or through the buffers is marginal as can be deduced by the large amount of buffer and small water flowrates. This applies also the chemical erosion of bentonite. The mutual degradation of the concrete and bentonite cannot be disregarded if these barriers are thin. In the examples 2 m thick barriers were used. The results suggest that even over a hundred-thousand year period, up to a metre of concrete and bentonite could be considerable degraded. Although the simple model tries to capture the main driving force for the chemical reactions there are considerable uncertainties in such modelling. It would therefore be prudent to assume that cement such as ordinary Portland cement should be avoided, pending more detailed investigations. The use of low pH cement seems to be considerably less prone to cause major concrete and bentonite degradation.

10 Discussion and conclusions

In the comparison of the different repository concepts simplified geometries were used in order to facilitate the analysis. Some simple models were developed that focus on the most important mechanisms and processes governing the nuclide release. The simple models were validated by more elaborate models, which needed to be solved by numerical methods. We deem the simple models to be sufficient and to give good insights into the main processes that determine the nuclide release.

The hydraulic cage was found to actually promote release of nuclides although it decreases the flow through the waste. For the tunnel with buffer a non-altered concrete buffer allows little water to flow both through the tunnel and the Supersilo design. Radionuclide release by diffusion will be larger than by flow with a non-altered concrete barrier. Bentonite, being much more permeable to flow, allows larger water flowrates through the waste. Release by flow can then under some circumstances be larger than by diffusion. Concrete allows considerably larger decay of the sorbing nuclides than bentonite even if the concrete has been mildly degraded. However, if the concrete develops highly permeable fractures this can lead to considerably increased releases in some situations, especially for the tunnel design. The Supersilo has some advantages over the tunnel alternative in that would be easier to avoid intersection with a fracture zone, which otherwise can be problematic for tunnels under some circumstances. In the Supersilo design option, with bentonite between waste and concrete wall, the bentonite will be subject to chemical degradation from concrete on both sides. Unless low pH cement is used the bentonite and the silo wall can be degraded to depths of up to a metre over 100,000 years. One may consider placing bentonite outside the thick-walled concrete silo instead. Then, even if the concrete wall should be severely fractured it would be protected from much increased flow by the bentonite and it would retain its sorption properties. The latter would ensure that the nuclides decay as in a non-degraded concrete. In the analysis the waste has been assumed to have much higher hydraulic conductivity than rock, bentonite and concrete. If it were to have much impact on the release it would have to have lower conductivity than the rock.

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