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# Progress report on evaluation of long term safety of proposed SFL concepts

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# Preface

This report is a result of a collaborative effort in the SFL Concept study, performed at SKB between 2011 and 2013. The author has summarized the work performed by the project group: Mattias Elfwing, Per Mårtensson, Pär Grahm, Mikael Gontier, Sofie Tunbrant and Lena Z Evins. In addition, this report has benefited from the reasoning and contributions from the following experts: Patrik Sellin, Ignasi Puigdomenech, Björn Gylling, Ulrik Kautsky, and Jens-Ove Näslund.

# Abstract

This report aims to summarize work performed in the project SFL Concept study, carried out at SKB between 2011 and 2013. Different aspects of long-term safety are discussed and a preliminary evaluation of the proposed concepts is presented. The concepts that are subjected to the evaluation are described in a general manner and they are in no way optimized with respect to performance. There are, at the time of writing, still some major uncertainties surrounding the long-lived waste, both in terms of inventory and waste form. In addition, no site is selected. Optimization of the repository design will therefore follow in later stages of the development of the design, and be linked both with improved waste characterization and the results of future safety assessments. The purpose of the current report is two-fold: to aid the choice of concepts to be further developed and analysed, and to provide a starting point for further research efforts required for a future safety assessment.

Requirements from regulations have guided the formulation of evaluation factors, which are used as a framework around which the studies and arguments are connected. For long-term safety, two evaluation factors have been formulated:

- 1) Feasibility of making a post-closure safety assessment.
- 2) Robustness of the barrier safety functions.

The proposed concepts are based around the idea of retarding the radionuclide release from the waste by surrounding the waste by different barrier materials in a geological repository placed 300 to 500 meters deep in crystalline bedrock. The different materials suggested are gravel, concrete, and bentonite. One concept combines all three of these materials, which suggests increased retardation potential, but also negative effects such as a possible detrimental interaction between the barrier materials. Also described are potential waste conditioning options, as well as a review of possible materials which may enhance sorption of some key radionuclides.

The safety functions are based on the properties of the materials in terms of hydraulic conductivity, diffusivity, sorption and other chemical properties. Thus, processes that might influence these properties are identified and the potential effects, during the time frame required (at least 100,000 years) are evaluated. In light of the first evaluation factor, a brief appraisal of the current process understanding, as well as the potential to define, achieve and verify the initial state is provided. The second evaluation factor involves an estimate of performance in the long-term, focused around studies and arguments concerning the evolution of the safety functions. Processes in the natural barrier (the bedrock), as well as other features and processes in the natural system, are expected to affect the engineered barriers. The effect of the natural barrier therefore plays a part in the preliminary evaluation. Research efforts needed in this area for a future safety assessment are identified. Some work has already been initiated; however, this work is mainly connected to later stages in the development of the SFL repository. Nevertheless, a brief overview of important aspects to consider in this area for future research is provided here.

Expressions for simple calculations for equivalent flow, i.e. the solute carrying capacity of water seeping in the rock surrounding a repository, is used in a comparison between the concepts. The results show that intact and pristine concrete is superior to clay when it comes to the retention of radionuclides; however, radionuclide transport will be dominated by diffusion in both, even in situations when the waste has a high hydraulic conductivity. Finally, using the evaluation factors as a guide, and the studies and arguments presented in the report, the preliminary evaluation suggests that the Concrete repository and the Clay repository are the most promising concepts. The Gravel repository is not suitable without further added safety functions. Using a combination of these barrier materials involves potential problems due to both concrete-bentonite interaction, as well as uncertainties concerning the initial state.

# Sammanfattning

Denna rapport syftar till att sammanfatta arbete som utförts i projektet SFL Konceptstudie, vilket genomfördes vid SKB mellan 2011 och 2013. Olika aspekter av långsiktig säkerhet diskuteras och en preliminär bedömning av de föreslagna begreppen presenteras. De koncept som utvärderas här beskrivs på ett allmänt sätt och de är inte på något sätt optimerade. Det finns, i skrivande stund, fortfarande några stora osäkerheter kring det långlivade avfallet, både vad gäller radionuklidinventarium och avfallsformer. Dessutom återstår platsval för SFL. Optimering av förvarets utformning kommer därför att följa i senare stadier av utvecklingen av förvaret, och kopplas både med förbättrad avfallskarakterisering och med resultaten av kommande säkerhetsanalyser. Syftet med denna rapport är dubbelt: att underlätta valet av förvarskoncept som ska vidareutvecklas och analyseras, samt att ge en utgångspunkt för den ytterligare forskning som krävs för en framtida säkerhetsanalys.

Krav från föreskrifter har styrt utformningen av de utvärderingsfaktorer, som används som ett stöd för de beskrivna undersökningar och argumenten. För den långsiktiga säkerheten har två utvärderingsfaktorer formulerats:

- 1) Möjligheten att genomföra en säkerhetsanalys.
- 2) Robusthet hos barriärernas säkerhetsfunktioner.

De föreslagna koncepten baseras kring idén om att retardera radionuklidutsläpp från avfallet genom att omge avfallet med olika barriärer i ett geologiskt slutförvar, vilket placeras på 300–500 meters djup i urberget. De olika föreslagna barriärmaterialen är grus, betong och bentonit. Ett koncept kombinerar alla tre av dessa material, vilket tyder på ökad retardationspotential, men också negativa effekter såsom en skadlig växelverkan mellan barriärmaterial. Möjliga konditioneringsmetoder beskrivs också, samt en översikt av material med potential att förbättra sorption av vissa nyckelnuklider.

Säkerhetsfunktionerna är baserade på hydraulisk konduktivitet, diffusivitet, sorption och andra egenskaper hos dessa material. Processer som kan påverka dessa egenskaper identifieras och de potentiella effekterna av dessa under den föreskrivna tidsramen (minst 100 000 år) utvärderas. I ljuset av den första utvärderingsfaktorn ges en kort bedömning av den aktuella processförståelsen, liksom möjligheten att definiera, uppnå och kontrollera initialtillståndet. Den andra utvärderingsfaktorn innebär en uppskattning av funktion på lång sikt, där fokus ligger på studier och argument avseende utvecklingen av säkerhetsfunktionerna. Processer i den naturliga barriären (berggrunden), samt andra funktioner och processer i det naturliga systemet, kommer att påverka de tekniska barriärerna. Och spelar därför en viss roll i den preliminära utvärderingen. Forskningsinsatser har identifierats, och visst arbete har redan inletts, men detta arbete främst kopplat till senare skeden i utvecklingen av SFL-förvaret. En kort översikt ges här över viktiga aspekter att beakta för framtida forskning inom dessa områden.

Uttryck för enkla beräkningar för ekvivalenta flödet, dvs. vattnets transportkapacitet, används i en jämförelse mellan koncepten. Resultaten visar att intakt betong är bättre än bentonitlera när det gäller radionuklidretention, men i båda fallen kommer radionuklidtransporten att domineras av diffusion. Detta gäller även när avfallet har en hög hydraulisk konduktivitet. Slutligen, med ledning av utvärderingsfaktorerna, samt de undersökningar och argument som presenteras i rapporten, tyder den preliminära utvärderingen på att Betong-förvaret och Bentonit-förvaret är de mest lovande koncepten. Grus-förvaret är inte lämpligt utan ytterligare säkerhetsfunktioner. Ett förvar som kombinerar dessa barriärmaterial innebär potentiella problem, på grund av interaktionen mellan betong och bentonit interaktion samt osäkerheter rörande initialtillstånd.

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# 1 Introduction

### 1.1 Background – the current and previous SFL concepts

The project SFL Concept study, carried out at SKB between 2011 and 2013, is a first step to constrain possible repository concepts for long-lived low and intermediate level waste. The goal is to choose maximum two different concepts for continued study and future evaluation using safety assessment methodology. The background to these studies is found in the 1999 safety assessment of SFL 3–5 (SKB 1999) and the reference inventory used for that assessment (SKBdoc 1416968). It was noted in that previous assessment that mainly two factors were important for the calculated result. Both of these factors were site dependant (hydrogeological conditions and biosphere conditions). Thus, the suggestion from the previous preliminary assessment was to reduce the release to the far-field by improving the engineered barriers. The repository design should be modified so that the barriers are more effective for reducing the release of the dose-dominant radionuclides: for the SFL waste, these were, in the 1999 study, mainly Cl-36 and Mo-93 (section 10-3 in SKB 1999).

### 1.2 Availability of updated information

There are two main types of waste from which the source term is derived: one type of waste already exists and is in storage on the Studsvik site (so-called "legacy waste", which includes waste from the early Swedish nuclear research programmes), while most of the other main type (metallic parts from the nuclear reactors) is expected to accumulate in the future. Thus, the 1998 waste inventory was based both on knowledge of existing waste, and forecasts. The radionuclide inventory was calculated from this, using correlation factors (SKBdoc 1416968). In parallel with the SFL Concept study, the SFL Reference inventory from 1998 is updated. The idea is to have an updated inventory for the next phase of deeper evaluation of the chosen concept(s). This new inventory is reported elsewhere (Herschend 2013). However, it should be noted that the legacy waste is, in its present state, complex, containing various types of materials. This waste may require further treatment and conditioning in order to agree with acceptance criteria. Thus, for the present report, the Reference inventory from 1998 is considered.

### 1.3 A note on uncertainties and the iterative process

Clearly, there are still some major uncertainties surrounding the SFL waste. This has implications for choices of waste conditioning and repository design of the engineered barriers. Therefore, if and when there is new information, it should be possible to go back and adjust the suggested concept appropriately, if deemed necessary. The changed concept should then be assessed in a similar manner; this iterative process is inherent in the process of designing a repository with long-term safety in focus.

### 1.4 Purpose and contents of this report

The purpose of this report is mainly to summarize the work performed regarding aspects of long-term safety, on which the evaluation of the proposed SFL repository concepts is based. In addition, this report is expected to function as a starting point for the more elaborate evaluations and assessments of the chosen concepts that are planned for the future. To reach these goals, this report describes the progress regarding the evaluation of long-term safety of different repository concepts suggested for the SFL Concept study. Information is provided on identified issues of concern for the long-term safety of a future SFL repository. In addition, the identification of such issues triggers further investigations and analysis, in order to aid the evaluation of the proposed concepts. Therefore, this report describes both the results of preliminary studies as well as suggestions for future research programs in different areas.

## 2 Method of evaluation and evaluation factors

The evaluation of the proposed concepts with regards to post-closure safety is strongly connected to estimates of how well the chosen barriers will function together. In this chapter, certain basic concepts are presented and discussed, followed by a description of the approach used here to perform the evaluation. The present evaluation represents the first steps in a more long-term endeavour, based on research to enhance process understanding and modelling approaches. As understanding proceeds, this is expected to provide feed-back to the technical aspects of the proposed concepts and repository designs.

### 2.1 A background to safety assessments

The complexity of the task to evaluate the long-term safety of a proposed repository is significant. The safety assessment methodology followed by SKB for a spent nuclear fuel repository is documented in SKB (2011) and a similar methodology will be followed in coming assessments of the SFL repository. A safety assessment requires knowledge about the initial state of the waste and the repository, and the natural system surrounding it. For all parts of the system, it is also required to evaluate the effects of all expected processes that can alter the system in the time frame required for compliance with the regulations. The time frame for the safety assessment of a repository containing long-lived radionuclides needs to cover at least 100,000 years, or the time for a glacial cycle, and it should not cover more than 1 million years (General advice to SSMFS 2008:37). This means that the time frame is comparable to that of the spent nuclear fuel repository.

Due to the radioactive decay of the waste, the activity will decrease at a certain rate, depending on the radionuclide inventory. It is important to note the differences in decay time between the actinides, and the shorter-lived fission and activation products. The SFL waste contains both shorter lived radionuclides, such as Ni-63 (with half-life of 96 years), but also actinides and some longer lived activation products, such as Cl-36 (with a half-life of ca 300,000 years). Thus, the future safety assessment of the SFL repository is required to provide quantitative measures of risk for the time period of at least 100,000 years. However, the time following 100,000 years up to 1 million years, also need to be addressed, at least in a qualitative way. This means, that the future safety assessment should take into account all processes likely to affect the repository system during the one million years following closure.

What is required is thus an evaluation of system evolution in a one million year perspective encompassing changes in climate, biosphere, surface system, bedrock, geochemistry, hydrology, engineered barriers, and waste. A safety assessment combines the most recent knowledge and development in all these areas, and extracts quantitative data for use in modelling of the system. The result is quantitative measure of risk as given by the model.

It is also noteworthy that in the future safety assessment of SFL, the risk analysis needs to be most detailed for the first thousand years after repository closure. Available site-specific data and details regarding the early development of the repository need to be used as input. This requirement of a more detailed analysis of the first thousand years may be significant for SFL, depending on the possibility of slow radionuclide release already from the time of closure.

### 2.2 Safety functions

In SKB's safety assessment methodology (SKB 2011), the barriers of a repository system are assigned one or several safety functions. The two main safety functions of SKB's spent nuclear fuel repository are containment and retardation. If the safety of the repository is built primarily upon containment, this implies that the chosen barriers are expected to fully contain the radionuclides within the repository. Applying retardation as a safety function implies that the barriers (both engineered and natural) are expected to efficiently retard the radionuclide migration once they have escaped the container, so that the exposure to the surface system is delayed and dispersed in time.

A multi-barrier repository system is composed of different barriers each with its safety functions. A barrier safety function is defined as a role through which a repository component (barrier) contributes to safety (SKB 2011). An example is the bentonite buffer in the KBS-3 repository system; there are several safety functions provided by this barrier. One is protecting the waste container (in the KBS-3 case, the copper canister) from advective flow of groundwater. Another is filtering of colloids; this contributes to retardation of radionuclides. Following the methodology in SKB (2011), each safety function is associated with one or several quantitative indicators that show how well the safety function in question is upheld. In some cases also quantitative criteria can be defined such that when an indicator fulfils a criterion, the safety function is upheld. Due to the nature of the indicators, the corresponding criteria can be either quantitative or qualitative.

It follows from the above that in a safety assessment, it is necessary to demonstrate how safety is related to the safety functions of the barriers and how these safety functions are affected by different processes which occur over time.

For the present report, the importance of the defined safety functions of the proposed barriers is such that they will allow identification of relevant processes to study; the question whether a barrier will be detrimentally affected or not by a process is rooted in the definition of the safety function assigned to the barrier.

### 2.3 Application of evaluation factors

In the SFL Concept Study, the proposed concepts are evaluated and compared. The overall method applied in the project is described in Elfwing et al. (2013). The general idea is to provide a frame-work for a transparent comparison where the concept evaluation is based on the same factors. These factors are called the Evaluation factors (EF). These are derived from laws and regulations as well as other requirements (see Elfwing et al. 2013). For the case of long-term safety, two main evaluation factors have been defined:

- 1) Feasibility of making a post-closure safety assessment.
- 2) Robustness of the barrier safety functions.

These evaluation factors illustrate two separate, but related requirements. The first is the basic need to provide a safety assessment. Thus, the concept should not involve any part which cannot be analysed, or which presents severe limitations to the possibility of an analysis. Concepts are evaluated on basis of the ease of achieving and determining the initial state, as well as on maturity of process understanding and modelling. Secondly, the suggested barrier concepts need to involve safety functions which are sound and robust. Concepts may thus be evaluated on basis of (preliminary) estimates of performance of the barriers with regard to safety function. This evaluation factor relates to the requirements that the barrier system should contain, prevent or retard the dispersion of radionuclides, and that the barrier system should be durable, i.e. it needs to protect against harmful effects of radiation for so long that the repository with its waste does not cause harm to human health or the environment. Sub-factors included in the evaluation are thus the potential of the engineered barriers to provide and maintain the required hydrological, mechanical and chemical properties. The possible effects of processes in both waste and other barriers on a safety function should be evaluated. The durability of the barrier may also be affected by the repository environment and the chemical reactivity of the waste.

Therefore, the evaluation factors are closely related to the defined safety functions, which in turn are related to processes; these processes need to be understood and modelled. This study of the relevant processes and how they may or may not affect the defined safety functions are at the heart of continued research for concept evaluation. Here, the evaluation factors are applied in a general sense during the course of the current project, to guide the extended analysis.

# 3 Concept descriptions

### 3.1 The method for concept identification and development

The general idea behind the SFL Concept study was to provide a transparent and traceable documentation of the concept identification and selection process. The method is described in Chapter 3 of Elfwing et al. (2013), and involves identifying as many strategies and concepts as possible, and then eliminating them one by one on the basis of laws and regulations and other requirements. The strategies and concepts that were abandoned in this first screening are briefly described in section 7.1 in Elfwing et al. (2013). After the first screening, the subsequent selection process was divided in two steps. First, all concepts identified were subjected to a primary assessment (see section 8.1 in Elfwing et al. 2013) based on reasoning rooted in the evaluation factors. During this primary assessment, a number of concepts were deemed not viable for further study, while four concepts remained to be further analysed. The remaining four concepts are briefly described below, with an emphasis on engineered barriers and safety functions. For a more detailed description, see chapter 7 in Elfwing et al. (2013).

### 3.2 Candidates for expanded analysis

The descriptions below are to be considered as suggested concepts, and the dimensions and quantities are therefore preliminary and in no way optimized for performance.

#### 3.2.1 The Gravel repository

#### Concept

The concept involves placing waste in a rock vault, and using gravel or crushed rock as backfill around the waste. The gravel should have known grain size distribution, and the hydraulic conductivity for the material used shall be  $10^{-5}$  m/s or greater. The thickness of the gravel bed on each side of the waste containers is 2.5 m, the thickness of the gravel bed above the waste containers is 6 m, and the waste rests on a concrete base slab placed on top of a ca 1 m thick gravel bed (Figure 3-1).

#### **Engineered Barrier**

The one engineered barrier in this concept is the gravel backfill. This is intended to function as a hydraulic cage with a high hydraulic conductivity, which directs the water flow around the waste rather than through it.

#### Safety Function

The one safety function of the engineered barrier is high hydraulic conductivity in the gravel backfill.

This contributes to retardation by ensuring that radionuclide transport in the waste is dominated by diffusion, not advection. The safety function is based on the assumption that the slow diffusive transport will retard the release of radionuclides compared with release dominated by advective transport.



**Figure 3-1.** Cross-sectional view of the Gravel repository concept. Legend: 1) Theoretical tunnel contour. 2) Gravel. 3) Waste containers. 4) Concrete base slab. 5) Gravel or crushed rock. Approximate dimensions: A = 20 m, B = 17 m, C = 15 m, D = 1 m, E = 2.5 m, F = 5-10 m.

#### 3.2.2 The Concrete repository

#### Concept

The waste is here placed in a rock cavern and using concrete as backfill, a large concrete monolith is produced. The waste is surrounded by an operational structure, i.e. a framework in which the waste is placed, composed of 0.5 m thick concrete walls. The inner void is filled with gas-permeable grout, and the space between the concrete walls and the rock is filled with concrete with a thickness of ca 2 m. The top concrete backfill will have a thickness of 5-10 m (Figure 3-2).

#### **Engineered Barrier**

The one engineered barrier is concrete. The solid concrete monolith is expected to reduce inflow of groundwater into the vault, and provide an environment characterised by low diffusivity, strong sorption and high pH.

#### Safety Functions

The safety functions are all related to the properties of the concrete: low hydraulic conductivity, low diffusivity, strong sorption of radionuclides, and high pH environment. This contributes to retardation by ensuring that radionuclide transport in the barrier is dominated by diffusion, not advection. In addition, radionuclide transport through intact concrete is slow. Radionuclide release from the waste to the concrete will be slowed down due to the passivating effect of the high pH in the pore water.



**Figure 3-2.** Cross-sectional view of the Concrete repository. Legend: 1) Theoretical tunnel contour. 2) Concrete. 3) Grout. 4) Reinforced concrete (0.5 m). 5) Waste containers. 6) Concrete. Approximate dimensions: A = 20 m, B = 17 m, C = 16 m, D = 2 m, E = 2 m, F = 5-10 m.

#### 3.2.3 The Clay repository

#### Concept

This concept involves an operation disposal structure made of ca 0.5 m thick concrete walls (without assigned long-term safety function). The backfill, including the bottom, is made of bentonite blocks with a dry density of  $1,600-1,700 \text{ kg/m}^3$ . The total thickness of the bentonite surrounding the operational structure is 2 m, apart from the top, which is 3.5 m (Figure 3-3). The dome above the bentonite is filled with bentonite pellets with a dry density of  $\sim 1,000 \text{ kg/m}^3$ .

#### **Engineered Barrier**

The engineered barrier is bentonite. Completely filling the rock vault with bentonite is expected to enhance retention through low hydraulic conductivity, low diffusivity, strong sorption and filtering of colloids.

#### Safety Functions

The safety functions are all related to the properties of the bentonite: low hydraulic conductivity, low diffusivity, strong sorption of radionuclides, and efficient filtering of colloids. This contributes to retardation by ensuring that radionuclide transport in the barrier is dominated by diffusion, not advection. In addition, radionuclide transport through bentonite clay is slow. The retardation is enhanced especially for colloid-transported radionuclides due to efficient filtering effects.



**Figure 3-3.** Cross-sectional view of the Clay repository concept. Legend: 1) Theoretical tunnel contour. 2) Bentonite pellets. 3) Grout. 4) Concrete structure for the operating period (0.5 metre). 5) Granite pillars. 6) Waste containers. 7) Bentonite blocks. Approximate dimensions: A = 20 m, B = 17 m, C = 16 m, D = 2 m, E = 2 m, F = 3-4 m, G = 2-3 m.

#### 3.2.4 The Super silo

#### Concept

The waste is here placed in a silo with a cylindrical double concrete wall. The rock cavern is ca 40 m high and 35 m in diameter. The space between the concrete walls is filled with bentonite on the side and on the top (at closure), and with a mixture of sand and bentonite at the bottom. The silo is placed on a layer of gravel and the space between the silo walls and the rock is also filled by gravel. When the lid is placed on the top of the silo structure, the top of the cavern is backfilled with gravel (Figure 3-4). The Super silo concept involves three different engineered barrier materials with different functions, all contributing to the retardation of radionuclides.

#### **Engineered Barriers**

The structure is composed of the following system of engineered barriers:

- Inner concrete silo: Concrete, thickness 1 m.
- Liner: Bentonite, blocks in bottom and lid, pellets on the sides; thickness 1 m. The dry density of the block filled part will be ~1,500 kg/m<sup>3</sup> while the pellets filled part will have a dry density of ~1,000 kg/m<sup>3</sup>.
- Outer concrete silo: Reinforced concrete, thickness 1 m.
- Outer backfill: Gravel, thickness 2 m.
- Bottom: Drained gravel, thickness 2 m.
- Top: Gravel, thickness 5–10 m.

It should be noted that the details provided here are examples and could be adjusted for optimization.



**Figure 3-4.** Cross-sectional view of the Super silo concept. Legend: 1) Theoretical rock cavern contour. 2, 3, 4, 5) Gravel or crushed rock. 6) Reinforced concrete. 7) Bentonite blocks. 8) Concrete. 9) Concrete shaft walls (0.5 m). 10) Waste containers. Approximate dimensions: A = 33 m, B = 35 m, C = 2 m, D = 25 m, E = 5-10 m, F = 2 m, G = 1 m, H = 1 m, J = 1 m.

#### Safety Functions

The concrete and the bentonite both provide low hydraulic conductivity, low diffusivity, and strong sorption of radionuclides. The surrounding gravel is expected to divert groundwater flow by providing an outer zone of high hydraulic conductivity.

All of these safety functions contribute to the retardation of radionuclide transport by ensuring that radionuclide transport in the barrier is dominated by diffusion, not advection, and that the diffusive transport of radionuclides through the barriers are slow.

### 3.3 Conditioning methods and other additional features

In addition to the concepts listed above, a number of different conditioning methods have been identified. These conditioning methods are described in Chapter 5 in Elfwing et al. (2013). They have not been evaluated in the same manner as the repository concepts, but some have been further analysed in order to estimate feasibility and potential. In the further development of the SFL concepts, the chosen conditioning methods, if any, will affect the initial state considered in the safety assessment. At the current stage, however, the uncertainties are still deemed too large to warrant even a preliminary evaluation of suitability. This report provides some information that has emerged from a screening of possible condition methods. Those deemed particularly interesting in view of long-term safety, are mentioned below.

#### 3.3.1 Sealed container

Placing the waste in sealed containers (Pettersson 2013) would ensure containment of the radionuclides for as long as the container is intact. Adding the safety function Containment to the concept clearly has the potential of improving the long-term safety. However, method development concerning welding 100 mm thick steel sheets is required. It also needs to be shown that the welds are tight and do not impair the containment function of the sealed container.

#### 3.3.2 Melting of metallic waste

Activated metallic waste is the main source for a large portion of the SFL radionuclide inventory. One way to lower the release rate from this waste is to drastically reduce the surface volume. This can be done by melting. This conditioning method is described in section 5.1.5 of Elfwing et al. (2013) and is subjected to a more detailed analysis in Huutoniemi et al. (2012). The analysis indicates that there are challenges with regards to non-normal operation, and potential consequences of an accident.

#### 3.3.3 Vitrification

This conditioning method could be possible for the existing legacy waste, considering the mixed and complex properties of this waste category (e.g. Hyatt and James 2013), and depending on available methods and the risks involved in handling the materials. The benefit of reconditioning this waste lies in improved long-term safety and the improved knowledge of the initial state of the waste, which is a required starting point when performing a safety assessment. Vitrification of the waste could be done in facilities using plasma furnaces, such as the Zwilag facility (Shuey and Ottmer 2006), where either the complete, grouted drum could be processed, or any fraction of the waste, if it is crushed and sorted.

The characterisation of the waste form is clearly important, and an improvement in this respect for the legacy waste should be given thorough consideration, in order to follow the IAEA guidelines for radioactive waste control (IAEA 2007).

#### 3.3.4 Chemically improved retention

If a large amount of material with properties beneficial for chemical retention of radionuclides, for example a high sorption material, is added to the waste form or the surrounding structures, this has the potential of improving long-term safety by enhancing the safety function Retention. This conditioning method is described in section 5.5.1 in Elfwing et al. (2013), and potential high-sorption materials are further analysed in Krall (2012). The main findings of Krall (2012) are provided in the current report (section 4.4). The most problematic radionuclides are those which form anionic species in natural environments; thus, the idea is to find a suitable material which has the potential to enhance the sorption of anionic species.

## 4 Processes in the engineered barriers

In order to guide investigations and research efforts, a preliminary overview is given here of the processes deemed relevant in light of the materials and related safety functions (see section 3.2). Due to the early stage in concept development, the list of relevant processes to study is expected to be expanded when the studies have progressed further. The processes and research needs described below are limited to those considered most relevant to the proposed concepts. The research needs were identified through discussions within the group and expert judgements. Different types of radionuclide transport, being consequences of several fundamental processes (e.g. advective water flow, diffusion, colloid release), are not considered further here, but are discussed in Chapter 6.

As mentioned previously, the study of relevant processes affecting safety functions of the different barriers are at the heart of the evaluation of the barrier performance. The concepts described above are based either on one-material barriers (gravel, concrete, clay) or a combination of these (the Super silo).

### 4.1 Gravel barrier processes

The Gravel repository is based on the safety function high hydraulic conductivity of the gravel backfill, so that it functions as a hydraulic cage. The concept of a hydraulic cage is employed elsewhere, e.g. the Richard repository, Czech Republic (Haverkamp et al. 2010). When considering the processes expected in the gravel, it could be viewed as a border between the engineered repository and the natural barrier. Many processes expected to be important in the surrounding rock are also expected in the gravel barrier. The processes of most concern fall within the realm of hydrogeology and geochemistry; in fact, in many cases one needs to consider the close interplay between these two areas of research, and the emphasis of the continued research is therefore to strengthen the connection between hydrology and geochemistry.

Central processes in most parts of the repository system are dissolution and precipitation. These processes are also considered of primary importance for the long-term function of a hydraulic cage. It is expected that gravel originating from crushed rock will provide a large reactive surface area, causing chemical reactions to occur throughout the volume. Clogging of the gravel may occur, not only by chemical reactions, but also through physical processes. This will affect the hydraulic conductivity, most likely in an inhomogeneous manner, possibly causing channelling effects. The effect of clogging on the hydraulic system therefore requires evaluation.

Matrix diffusion, i.e. diffusion of groundwater solutes from water flowing in gravel and fractured rock into the more stagnant water in the rock matrix is expected to be important for SFL specifically. Many SFL-specific radionuclides are transported as anions, and do not sorb efficiently on surfaces. Taking this process into account requires modelling, and model development, using experimental data.

If gravel is to be used as a hydraulic cage, as in the Gravel repository, and the Super silo, research should be focused on first, testing the assumption that the gravel will in fact function as envisaged, and secondly, how the properties of the gravel will change over time.

### 4.2 Concrete barrier processes

The concrete repository is expected to provide the safety functions low hydraulic conductivity, low diffusivity, strong sorption of radionuclides, and high pH environment. There are a number of processes that will affect the concrete after repository closure, all contributing to concrete degradation and impairing the safety functions. The chemical alteration of the concrete matrix, involving dissolution of existing material and precipitation of secondary minerals are central processes in the inevitable, long-term degradation of concrete. These processes are closely connected with the physical process of fracturing. The changes of mineral composition of the concrete during degradation will also affect the sorption and speciation of radionuclides that are transported through the concrete.

Dissolution and precipitation will not only affect the chemical properties but also the hydraulic properties and water flow through the concrete will change as the concrete is degraded. Therefore, the balance between diffusive and advective transport of radionuclides through the concrete will be affected by the degradation. Research in this area is performed by SKB and internationally (Grandia et al. 2010, Savage et al. 2011), and it is expected that this will remain an area of research focus in the years to come.

Freezing of concrete will clearly accelerate the degradation processes. In the proposed SFL concepts, the repository is placed at depths between 300–500 m. One main reason for this is to avoid the freezing of the concrete structures. However, depending on which site is chosen, and details of the design, further investigations concerning different aspects of freezing will be required.

The pore water in the concrete structure will be alkaline. The pH will initially be very high, and slowly, in conjunction with concrete degradation, change to lower values. However, the massive amount of concrete which is envisaged in the Concrete repository is expected to ensure an alkaline environment for a very long-time after repository closure. The high pH suggests that a passivating layer is formed on metals in the waste; this passivating layer will reduce metal corrosion rates and limit radionuclide release from metallic waste. Thus, it will be important to study in detail the evolution of the chemical properties of the pore water. The interaction between concrete and ions in the solution, originating either primarily from the groundwater or from the degradation and dissolution of the waste, is also an important process to consider.

For the Concrete repository, the process understanding of relevant processes occurring in pristine concrete is sufficient for the modelling required in a safety assessment; the uncertainties lie in the verification of the initial state, and the degradation processes. Areas identified for further studies and research include development of alternative materials for concrete reinforcement, gas formation and transport in concrete, how material strength is affect by degradation, etc.

### 4.3 Bentonite barrier processes

Bentonite is the engineered barrier component of the Clay repository, and an important part of the Super silo barrier system. There are many processes relevant to the evaluation of the safety functions, which for the Clay repository are low hydraulic conductivity, low diffusivity, strong sorption of radionuclides, and efficient filtering of colloids.

The material properties and expected long-term evolution of bentonite is well known for a KBS-3 type repository. However, the proposed SFL concepts under discussion are very different, and how the different dimensions and interacting materials may affect the bentonite evolution is not yet fully investigated. The processes, for example swelling, that occur during water saturation of the bentonite are of particular interest. Thus, the transition between unsaturated and saturated phase needs to be understood and modelled, in order to evaluate the hydraulic conductivity and diffusivity. Water transport is expected to change during the saturation phase, and there are potential complications if the swelling is inhomogeneous. Gas transport is also of particular importance for SFL, if significant gas production from, for example, metal corrosion, is expected.

The effects of bentonite erosion, i.e. colloid release triggered by groundwater of low salinity, must be considered, as must the colloid transport through the bentonite. The function of a fully saturated bentonite is such that it will filter colloids efficiently; however, processes such as dissolution and precipitation may affect this safety function, as well as the sorption and ion exchange. Liquefaction and freezing may also affect the function of the bentonite should such conditions become relevant.

Swelling of the bentonite will also affect the waste packages and any operational structures created around the waste. For the Super silo, swelling must also be taken into account when designing the double concrete silo wall. Importantly, the interaction between concrete and bentonite is detrimental for both materials, and research illuminating how the different components will affect each other both chemically and mechanically is required. Here one needs to consider also the effect of the waste packages.

### 4.4 Chemically improved retention

It has been observed that sorption of radionuclides is an important process with direct influence on the overall retardation safety function. Therefore, the idea that sorption in the repository could be improved by the addition of some suitable material, has been investigated at depth in a literature review (Krall 2012). The aim is to evaluate the potential of this idea by reporting the existing information on sorption in relevant materials. The following is a summary of this evaluation.

The conditioning method involving addition of a "high sorption material", described in section 5.5.1 of Elfwing et al. (2013), could possibly be applied as an improvement to any of the repository concepts described in section 3.2 of the current report. By adding a large amount of material with the sought-after characteristics, the near-field release of radionuclides could be significantly reduced, mainly through the process of sorption. In order to develop this idea further, and evaluate the potential, this option was further investigated by Krall (2012). This literature review revolves around two central questions: 1) what radionuclides are the ones we expect to contribute most to the risk? 2) what materials are available which may function as a high-sorption barrier for the specified radionuclides?

Information from the previous safety assessment (SKB 1999) provided enough background in order to identify a list of relevant radionuclides; the review indicates specifically Cl-36, Mo-93, C-14, and I-129, all of which are expected to occur as anionic species in the repository and natural system. These anionic species do not exhibit strong sorption to many materials as opposed to cationic species, which sorb well to materials such as bentonite, concrete and zeolite. Krall (2012) identified the following materials as relevant for sorption of anions:

- Layered double hydroxides (LDHs).
- Hydrous metal oxides.
- Zero-valent iron/Iron minerals.
- Red Mud.

Some of these are amphoteric, meaning depending on chemical environment they can sorb either cations or anions (Krall 2012). Some materials are also mineralogically and chemically complex. Red Mud, for example, is a mixture of very fine-grained minerals found in a waste product derived from the processing of aluminium ore. The exact composition can be hard to identify exactly, but can be better controlled by choosing a specific chemical treatment. Aside from the complexity, the Red Mud has many other promising attributes. It should also be noted that the three first types of materials are closely related, and one mineral can belong to more than one class (e.g. fougerite: an LDH, in some instances called green rust, which forms during anoxic corrosion of zero-valent iron).

Below follows a summary of the characteristics of the anion-sorbing materials, based on the findings of Krall (2012).

#### Layered double hydroxides (LDHs)

These materials, described as hydrotalcite-like compounds, exhibit the ability to capture organic and inorganic anions through the process of adsorption and ion exchange. They do occur in nature (e.g. hydrotalcite) but are relatively rare; in contrast, they are quite straightforward to synthesize. The LDHs have a layered structure consisting of "brucite-like" (brucite:  $Mg(OH)_2$ ) sheets stacked on top of each other. The anions can be accommodated between the sheets, which commonly have a positive charge. The most common anion found in naturally occurring LDH is carbonate, but halides, oxyanions, silicates, etc can also be incorporated.

Experiments have been performed in order to establish how efficiently the LDHs can sorb some anions. One example is reported by Wang and Gao (2006), who synthesized a Ni-Al LDH with a Ni/Al ratio of 3:1; this exhibited the highest sorption capability for  $TcO_4^-$  (pertechnetate), with a K<sub>d</sub> value of 307 mL/g at a pH of 8. The suggestion is that molybdate (MoO<sub>4</sub><sup>2-</sup>), being smaller and more negatively charged than pertechnetate, may adsorb to the Ni-Al LDH more efficiently.

#### Hydrous metal oxides

Metal oxide surfaces (or any oxide surface) are usually covered with hydroxyl (OH) groups, attached to the surface by dissociative chemisorption of water to the surface. The OH-groups enable the oxide to function as either an anion or cation exchanger, depending upon solution pH (these materials are thus amphoteric). This surface cover of OH-groups mean that during precipitation, when there is a build up of the fresh precipitates which are poorly crystalline and occur as very small particles, they will contain loosely bound water molecules in their structures. As these precipitates age, they become more crystalline (Dyer 2000).

Well known examples of these types of materials are trivalent hydrous oxides of iron and aluminium:  $\alpha$ -FeOOH (goethite),  $\beta$ -FeOOH, and  $\gamma$ -FeOOH (lepidicrocite),  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (corundum),  $\alpha$ -AlOOH (diaspore), and  $\alpha$ -Al(OH)<sub>3</sub> (gibbsite). Other hydrous oxides exhibiting similar ion exchange properties are those of Mn, Si, Sn, Ti, Zr and Th. Hydrous tin oxide may be most interesting for uptake of anionic species, as is indicated by the results of White and Rautiu (1997) which show uptake of IO<sub>3</sub><sup>-</sup> at a wide range of pH.

#### Zero-valent iron (ZVI) and iron minerals

Iron is a redox-active element and its behaviour as a function of Eh and pH is both complex and important for modelling chemical reactions. Pure iron and steel, both zero-valent iron (ZVI) materials, are relatively common in repository settings, and iron and its corrosion products have been investigated for the purpose of enhancing our understanding of radionuclide sorption onto these materials. Remediation of groundwater is another area where these materials are of specific interest (see for example Hashim et al. 2011). As applied to groundwater remediation, ZVI has been found to be a strong chemical reductant, with the ability to convert many mobile oxidized oxyanions (e.g.  $\text{CrO}_4^{2-}$ ,  $\text{TcO}_4^{-}$ ,  $\text{MOO}_2^{2-}$ ,  $\text{AsO}_4^{3-}$ , and  $\text{SeO}_4^{2-}$ ) and oxycations (e.g.  $\text{UO}_2^{2+}$ ) into immobile forms.

It appears as if in most cases, the function of the ZVI materials is stepwise, involving an initial redox reaction whereby ions in solution are reduced, and the reduced forms are then either sorbed onto the Fe-corrosion products, or precipitated as a solid phase. Therefore, sorption occurs mainly on the various secondary Fe(II)- and Fe(III)-minerals that may form on the ZVI surface. It should be noted that one corrosion product in particular, called green rust (fougerite), is a type of LDH and thus functions as such, and is capable of intercalating anions (see above).

#### Red Mud

As mentioned above, Red Mud is a waste product derived from the digestion of bauxite, an aluminium ore. The material is at first very alkaline (pH 10–12.5); however, by treatment with either acid or seawater the pH may be lowered to more acceptable levels. The exact composition of the red mud produced depends on the particular chemical and mineralogical composition of the bauxite before digestion. Bauxite commonly consists of Al-oxides and hydroxides; examples of minerals are gibbsite Al(OH)<sub>3</sub>, boehmite AlO(OH), and diaspore AlO(OH). It may also contain sheet silicates (kaolin), quartz and various Ti- and Fe-oxides and hydroxides (e.g. hematite, goethite and anatase). After Al-ore processing, the untreated Red Mud contains a variety of minerals, such as the following:

- Cancrinite  $[Na_6Ca_{1.5}Al_6Si_6O_{24}(CO_3)_{1.6}]$ .
- Hematite [Fe<sub>2</sub>O<sub>3</sub>].
- Sodalite [Na<sub>8</sub>(Cl,OH)<sub>2</sub>|Al<sub>6</sub>Si<sub>6</sub>O<sub>24</sub>].
- Boehmite [AlO(OH)].
- Gibbsite [Al(OH)<sub>3</sub>].

Cancrinite is the primary mineral before and after acid treatment of Red Mud. It has an open porous structure and can be considered as a material with zeolite-like properties. Acid treatment will however lower the amount of cancrinite with respect to hematite, so that acid-treated Red Mud contains a higher percentage of hematite. One study observed that acid treatment with HCl decreased the red mud's capacity to adsorb the cations by 30% (Santona et al. 2006). Seawater neutralisation of Red Mud is considered by some parties a better option than treatment with strong acids (Palmer et al. 2009). The chemical reactions involved in sea-water neutralisation result in the formation of Mg-, Ca-, and Al-hydroxides and carbonates of Mg, Ca, and Al, such as hydrotalcite, an LDH. The efficiency of removal of anionic species by different types of hydrotal-cites (3:1 and 4:1 hydrotalcite) with respect to removal of molybdate, arsenate and vanadate shows that 3:1 hydrotalcite removed molybdate most efficiently, while the 4:1 hydrotalcite increased the percentage of all anions removed from solution in the same aqueous system (Palmer et al. 2010).

It should be noted that many of the minerals occurring in both untreated and treated Red Mud have been mentioned in the previous sections as relevant materials for sorption of anions. Red Mud has also been studied for the sake of removal of cationic  $Cs^+$  and  $Sr^{2+}$  (Apak et al. 1995) with the conclusion that this type of material may be utilized as a barrier around shallow-land burial sites of low-level radioactive wastes and heavy metal-containing products.

### 5 Processes in the natural system

The natural system is everything between the repository and the human exposed to risk. It encompasses the bedrock, which constitutes the natural barrier, the fractures in the rock and the groundwater flow and chemistry both in the deep bedrock and on the surface. Knowledge concerning surface hydrology, landscape evolution and ecology are essential in order to estimate the fate and effect of the radionuclides once they reach the surface. All these aspects of the natural system are connected to the climate and what scenarios are chosen to represent the future climate evolution.

#### 5.1 Geosphere processes

The crystalline bedrock between the repository and the surface system constitutes the natural barrier in all concepts described in section 3.2. The number of processes affecting the safety functions of the natural barrier is large. However, the features and processes in the bedrock are site-specific; at the time of writing, the site-selection process in not yet initiated. Thus, the current report does not include any overview of the relevant geological studies. In the future stages of concept development and site selection, however, studies will have to take into account the interplay between geology and repository design. One example is the shape and volume of the repository, and the likelihood of intersection with a fracture zone. In some cases, a silo could be the better alternative in this respect, while at other sites, a long horizontal vault may be more beneficial. The effect of reactivation and movement along a fracture zone may also differ, depending on the shape of the repository, the backfill material chosen, and the hydrogeology at the site.

Results of some on-going and planned research projects will be applied for SFL; these relate generally to improvement of the tools used in modelling the fracture network, the hydrogeology and reactive transport of elements. Finally, considerable information regarding relevant geosphere processes is provided by the latest safety assessment for the spent nuclear fuel repository (SKB 2010a). Many processes described there are expected to be relevant also for any future safety assessment of the SFL repository.

### 5.2 Climate and scenarios

Future climate changes are expected to affect all parts of the repository system. The way this is handled in the safety assessment is usually to set up cases with different types of possible future climate developments. The climate cases vary in terms of the temporal development and characteristics of climate, ice sheets, permafrost, sea level and denudation. Different types of engineered barriers respond differently to environmental changes caused by climate change. Depending on the choice of repository concept, a number of relevant climate cases need to be identified and described. The climate cases result in a certain landscape development, and together the different cases make out a framework for the different calculations performed in a safety assessment. The climate cases are central to the formulation, construction and analysis of safety assessment scenarios, and there is a need to coordinate the efforts made in the complete chain of models used in the areas of climate, landscape development, hydrogeology, hydrochemistry, radionuclide transport and dose calculations.

Much knowledge regarding the climate and its potential effect on a geologic repository is already in place from previous SKB safety assessments (SKB 2010b). However, some repository concept specific issues that require further study includes development of ice sheet and permafrost for climate cases including cold climate conditions, and studies of processes related to global warming for warm climate cases. In the former case, the engineered barriers will be affected by an increased hydrostatic pressure induced by an overlying ice sheet. Thus, it is important to establish the maximum possible ice sheet thickness. This value will vary depending on the geographical location of the site. Depending on the outcome of ongoing studies for the spent nuclear fuel repository, complementary numerical simulation of the ice sheet and climate for the penultimate glaciations (the Saalian), i.e. the largest glaciation of the Quaternary period, may be needed. Both concrete and bentonite may be affected negatively by freezing, and therefore the repository is to be placed at a depth where the risk for barrier freezing associated with permafrost development is negligible. Therefore, the development of permafrost needs to be understood and described in a climate case with deep permafrost. Since the expected maximum freezing depth varies according to many parameters, this need to be studied for various possible repository sites.

Two climate cases, Global warming and Extended global warming (SKB 2013), describe an extended future period of warm climate. This type of climate development may affect the ground-water chemistry through i) long-term meteoric groundwater recharge and ii) long-term chemical bedrock weathering under warm conditions. The altered groundwater chemistry may in turn affect repository barriers. Bentonite erosion may, for example, result from contact with ground water of low salinity. The effect of prolonged temperate climate conditions on groundwater chemistry is therefore an important area for further study.

### 5.3 The surface system and the biosphere

As mentioned above (section 5.2) it is recognised that for any future safety assessment of SFL, models of the different systems need to be coordinated. The central role of the biosphere studies is to provide means for establishing dose from radionuclides released from the repository. Before this can be done, it is important to understand the behaviour of key radionuclides in the surface system and biosphere. Many radionuclides relevant for SFL are not among the key radionuclides in the spent nuclear fuel repository. One example is Cl-36, a mobile radionuclide with a relatively long half-life. Knowledge regarding transport and accumulation of this radionuclide is of vital significance for the future safety assessment of the SFL repository. This and other radionuclides are transported mainly as anions that do not sorb well on mineral surfaces. The reactivity of chloride needs to be revised, however, since it has been shown to be more reactive than previously thought. Recent studies show that the amount of organically bound chlorine is higher than that of mobile chlorine, which might be due to biological processes in soil. This probably affects the retention of chlorine (Bastviken et al. 2013).

Biological processes are likely also to affect the distribution and speciation of other important biologically active elements in the SFL waste, such as radium (Ra), carbon (C), iodine (I), nickel (Ni) and molybdenum (Mo). Calculating the dose from released radionuclides of these elements require research efforts. The transport and behaviour of these key radionuclides need to be modelled; however, the method, including the models it will use, needs to be developed and tested, and the results need to be summarised and evaluated.

In the safety assessment SR-Site (SKB 2011), the calculations were performed with Landscape Dose Conversion Factors (LDF), a radionuclide-specific dose conversion factor relating radionuclide release to dose. This factor does not change with time or location. It is anticipated that this treatment may be too simplified for the SFL-specific radionuclides. The calculation of dose may have to be more firmly connected to when and where the radionuclides are released, which means that the different models describing the repository, the geosphere and the biosphere need to be connected. Synchronising landscape and climate development will allow this connection.

6

### Preliminary assessments of near-field transport capacity

The basic variants of the concepts proposed are all based on retardation as the main safety function. A sealed metal container is suggested as a possible conditioning method (Pettersson 2013), which would provide an additional barrier with a containing safety function. However, it is advisable to investigate the necessity of such a barrier before developing it further. Thus, a comparison between the basic concepts, involving no containing function of the waste package, was performed (Neretnieks and Moreno 2013).

Radionuclides can be transported both by the water that flows *through* the waste and buffer (denoted Q), as well as diffusion through the buffer to the water that flows *past* the buffer, or in the case of the hydraulic cage, past the waste. The transport capacity by the latter mechanism is determined by the equivalent flowrate (denoted Qeq).

The following text is a summary of the comparison of near-field transport capacities of the proposed repository designs using the Qeq concept (Neretnieks and Moreno 2013).

### 6.1 Description of the conceptual models

Two main options are explored; using a hydraulic cage, with advective flow around the waste, and using a buffer as a diffusion barrier, i.e., ensuring transport by diffusion around the waste.

#### Basic data and conceptual models

In the *Gravel repository*, the waste containers are surrounded on all sides by gravel that acts as a hydraulic cage. The concept relies on minimised flow through the waste which is expected as long as the gravel cage can divert the flow. Nuclides will escape partly by flow through the waste and partly by diffusion from the waste to the water flowing in the gravel. In the case of a well-functioning hydraulic cage, nuclide escape by diffusion will dominate.

In the *Concrete repository* the waste containers are surrounded by concrete on all sides. The concrete is expected to act like a diffusion barrier, provided that neither significant fracturing nor chemical alteration has degraded the concrete. Should eventually concrete and waste inside it become more permeable than the surrounding rock, water flow will be directed to pass through the barrier and waste, rather than around it. However, strong sorption may still be active for some radionuclides, and pH will be maintained above 12 for a very long time if Portland cement is used.

The *Clay repository* is relying on the function of a thick bentonite barrier surrounding the waste on all sides. As long as the bentonite is less permeable than the surrounding rock nuclide escape will be dominated by diffusion through it, and some radionuclides can be expected to sorb to the bentonite. Closest to the waste, which itself contains concrete and is placed in a concrete framework, the bentonite may degrade by reactions with hydroxyl ions diffusing out from the concrete. Bentonite is also sensitive to low salinity of the groundwater.

The *Super silo* is 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. The concept is built on the idea that the multibarrier system will ensure that the release of nuclides to the seeping water in the rock is only by molecular diffusion. Sorption will be efficient for many radionuclides in both the bentonite and the concrete.

Basic data required for the modelling of the systems are: density, porosity, sorption coefficients (for Cl-36, Ni-59, and Ra-226) retardation factors, and half-lives (for Cl-36, Ni-59, and Ra-226). Data concerning flow and diffusion are: Hydraulic conductivity, effective diffusion coefficient, porosity, pore diffusion coefficient, and buffer thickness.

The calculations aim to provide estimates of the carrying capacity of nuclides by the water flowing in and around a repository vault. The calculations use constant release rate, or constant sorption coefficients for instantly dissolved nuclides. Molecular diffusion will dominate when the advective carrying capacity is small, as is expected when the waste is surrounded by low permeability barriers such as bentonite or concrete. The diffusivity then determines the rate of radionuclide escape to the seeping water in the rock, and the system can be considered as a series of transport resistances (as an electrical circuit). For advective flow, the carrying capacity is essentially the flowrate itself, while for diffusion, an equivalent flowrate (Qeq) is defined for the estimation of carrying capacity. Using the approximation that the diffusion coefficient in water is the same for all solutes, the equivalent flowrate only depends on the properties of the rock (the Q-equivalent concept is described on pages 18–19 in Neretnieks and Moreno (2013)).

#### Model limitations and simplifications

The simplification that is inherent in the calculations and modelling presented here should be noted. For example, the hydraulic and diffusive properties of the rock, hydraulic cage, buffer and waste are given a few discrete values in a base case. Since no site is yet selected for SFL, the hydraulic data of the site in the models are largely based on conditions at the Laxemar site. The treatment of the fracture network is as a porous medium rather than a more realistic fracture network.

Regarding radionuclide release from the waste form, there are many uncertainties. In the scoping calculations presented here, only constant release rate from the waste form or constant sorption coefficients for instantly dissolved nuclides is considered. For the hydraulic cage, the model considers the gravel a high conductivity region surrounding a low conductivity waste; the escape of radionuclides depends on diffusion through the waste and the direction and magnitude of flow around the waste. Besides the waste containers, the gravel is the only protective barrier aimed to divert the seeping groundwater around the waste to minimise flow through the waste.

The scoping calculations involve initial, non-degraded barrier materials, and the comparisons are valid for situations where the concrete and the bentonite have retained their low conductivity for a very long time.

### 6.2 The hydraulic cage

In the case of a hydraulic cage, there will be advective flow around the waste, and radionuclides will be released to the flowing water at the rate of diffusion out from the waste. The scoping calculations show that the flowing groundwater will, fairly rapidly, be equilibrated with regards to radionuclide concentration. This means that the concept of only using gravel around the waste is not efficient in retarding the release of radionuclides (Neretnieks and Moreno, page 24).

### 6.3 Diffusion dominated transport through buffer

A simple tool is used to estimate and compare how the bentonite and concrete barriers affect the release of nuclides from the barrier. By means of this simple tool, a fast impression of the retention capabilities of the buffer is given, as well as an indication of the impact for different radionuclides of the thickness, diffusion and retardation properties of the buffers. Sample calculations performed with Cl-36, Ni-59, and Ra-226, show that when the barriers are intact, sorbing nuclides decay considerably before they reach the seeping water in the rock. The estimated radionuclide release is increased if the hydraulic conductivity of the rock is higher.

The results show that bentonite and especially concrete cause many sorbing nuclides to decay to insignificance during their passage through the buffer. If the low conductivity is retained over long times, the concrete barrier is superior to the bentonite in the extent to which nuclides can decay during their passage. A more complex model, involving diffusion in the waste, buffer and the seeping water in the rock, has been used to test the analytical solution, and to estimate radionuclide retention in the barriers. Both the simple, analytical approach and the numerical model show that,

under diffusion-dominated transport conditions, concrete retards nuclides considerably better than bentonite. The results indicate that the use of the simplified model is appropriate for scoping calculations of radionuclide release rates, allowing the provisional comparison of alternative repository concepts.

### 6.4 Transport by flow through the waste

If the waste form is highly hydraulically conductive, and is embedded in a low conductivity buffer, it may result in considerably larger flows than if waste and buffer have similar conductivities. Simple formulas describing flow both parallel and perpendicular to the vault are used to assess impact for flow through the waste. Radionuclides will be transported both by advective flow and simultaneous diffusion through the buffer.

In this situation, the transport through the buffer(s) is still dominated by diffusion, and only marginally influenced by flow in the buffer or in the waste. Many sorbing nuclides will be retarded and decay to insignificance during their passage through the buffer. One or more highly transmissive fractures through the buffer would change this situation, however, and the nuclides would not be retarded by sorption but would escape with the flowing water without much decay. Fracturing is expected to be more likely for concrete than for bentonite, indicating that this situation is more likely to occur in the concrete barrier.

### 6.5 Degradation of concrete and bentonite

It is noted that long-term interaction between concrete and bentonite may cause the loss of desirable properties and impair the associated safety functions. The concept which is most vulnerable for this is the Super silo; however also the Clay repository involves some concrete operational structures. A simple model is presented by Neretnieks and Moreno (2013), which describes the rapid, solubility-limited dissolution of calcium hydroxide from the concrete, and its rate of diffusion to the concrete-bentonite interface. This causes alteration of the smectite, which turns into non-swelling minerals. Using conventional Portland cement can lead to a degradation up to a meter or more over a time period of 100,000 years. The use of low-pH cement is suggested in order to limit the damaging concrete-bentonite interaction.

### 6.6 Comparisons based on the preliminary results

The different concepts have been compared using calculations involving properties of rock, buffer and waste. Table 6-1 provides some basic data for buffer materials, as well as the parameter H, which is used to assess how efficient a buffer is in retarding a specific radionuclide. It can be seen from this table alone that the retention properties of pristine concrete is better than for clay (bentonite). The calculations indicate that the flow through the gravel repository is very high, as expected. However, the protecting effect of the hydraulic cage is not manifested in this case, due to diffusional radionuclide release from the waste to the water flowing in the gravel. In this case, the water in the gravel will be rapidly (in about 250 years) equilibrated with the waste, and any further calculations relating to the gravel repository are therefore abandoned.

For a preliminary evaluation of radionuclide transport away from the repository, the equivalent flow (Qeq), which is only dependent on rock properties, is coupled in a simple model with the flow in the vault and radionuclide concentration at the buffer-rock interface. See Table 6-2 for a summary of results. A few general observations are made: 1) the shape of the vault, here specifically tunnel or silo, affects Qeq; 2) the direction of the regional hydraulic gradient affects both Q and Qeq; 3) if the waste has high conductivity, the flow rate becomes higher. The worst case is when there is a hydraulic gradient which is parallel to a vault filled with bentonite. In all cases, however, diffusion dominates the transport (Peclet numbers below 1).

Table 6-1. Data from Tables 4-2 and 6-1 in Neretnieks and Moreno (2013) influencing radionuclide retention estimated from the preliminary calculations. The larger the H-value, the more efficient is the radionuclide retarded in relation to the decay constant of the radionuclide. N.d. = Not determined.

Data (pristine barrier)	Gravel	Concrete	Clay
Porosity (ε)	0.3	0.4	0.15
Hydraulic conductivity (Kh)	3.00E-06	5.00E-12	1.00E-10
Pore diffusion coefficient (Dp)	1.67E-09	6.67E-11	2.50E-10
H(C-14)	N.d.	26	0
H(CI-36)	N.d.	0	0
H(Ni-59)	N.d.	3	1
H(Ra-226)	N.d.	25	2
H(Pu-239)	N.d.	63	8

Table 6-2. Calculated flow rates and Peclet numbers for the different concepts, from Tables 5-2 and 7-2 in Neretnieks and Moreno (2013). A: waste with low hydraulic conductivity. B: waste with high hydraulic conductivity. Peclet number below 1 indicates that radionuclide transport is dominated by diffusion. N.d. = Not determined.

Parameters	Concrete		Clay		Super silo (Concrete)	Super silo (Clay)
Hydraulic gradient	parallel	perpendicular	parallel	perpendicular		
Equivalent flow (Qeq) in rock (m <sup>3</sup> /yr)	0.023	0.075	0.023	0.075	0.041	0.041
Flowrate (Q) through vault (A)	1.20E-04	3.70E-03	2.40E-03	6.90E-02	N.d.	N.d.
Flowrate through waste (B)	5.50E-03	5.50E-03	1.06E+00	1.10E–01	4.60E-03	9.30E-02
Peclet number (B)	0.16	0.011	0.32	0.023	0.029	0.058

The following section summarizes the comparisons presented in Chapter 9 of Neretnieks and Moreno (2013).

#### Gravel repository

The flow rate in the hydraulic cage is governed by the hydraulic conductivity of the surrounding rock which includes effects of fractures intersecting the vault. The preliminary results show that the hydraulic cage, as described in the base case, does not contribute to retardation. Therefore, the gravel repository is not considered a good option.

#### Concrete repository

The barrier considered in this case consist of non-degraded and unfractured concrete; this permits very little flow through the vault. The radionuclide release will be governed by the equivalent flow rate, which is ca.  $0.02 \text{ m}^3/\text{yr}$ . This number will increase if a prominent fracture or fracture zone intersects the vault.

The transport through the pristine concrete will, in all calculated cases, be by diffusion. However, it is noted that for the concrete repository, fracturing of the concrete would change the flowrate through the buffer and waste. A special concern is the possible situation where a transmissive fracture in the concrete is adjacent to a prominent fracture in the rock, which can result in a total flowrate on the order of  $1 \text{ m}^3/\text{yr}$  through the waste. Intact concrete has properties that are very beneficial for radionuclide retention; diffusion will dominate and many of the strongly sorbing nuclides will decay to insignificance in the concrete. It is estimated that even if the concrete that has to some degree degraded so that the pore diffusion coefficient has increased tenfold, the retardation properties are better than in bentonite.

#### Clay repository

The equivalent flow rate is the same as for concrete, about  $0.02 \text{ m}^3/\text{yr}$ . The flow through the vault is however different, since according to the base case, bentonite has a higher hydraulic conductivity than intact concrete (see Table 6-1). Many actinides and fission products move very slowly through the bentonite, but overall radionuclide retention is not as strong as for intact concrete. Due to material characteristics, bentonite is judged to be less prone to develop fractures than concrete. Concrete present in the waste and in operational structures will have a negative effect on the bentonite, as will very diluted groundwater.

#### Super silo

Due to the negative effects of the concrete on bentonite, the Super silo concept is sensitive to details in the design. If the bentonite is placed in between concrete structures, as is envisaged in the doublewalled Super silo concept, the bentonite will be attacked from both sides. Water may flow in fractures in the outer concrete silo wall directly to the bentonite and short-circuit the diffusion barrier in the concrete. If, however, the design is changed so that bentonite is placed outside a concrete silo wall, the bentonite may limit the flow rate in to the concrete. It is envisaged that this arrangement will also lessen the effect on radionuclide retardation by fracturing in the concrete. The main concern for the silo, involving both bentonite and concrete, is that the direct contact between concrete and bentonite will promote degradation of both buffers.

# 7 Evaluation of concepts according to evaluation factors

As was described in section 2.3, there are two main evaluation factors that are here considered for the guidance of the evaluation of the concepts with regards to the long-term safety.

These are:

- 1) Feasibility of making a post-closure safety assessment.
- 2) Robustness of the barrier safety functions.

They connect to requirements as described in Chapter 3 of Elfwing et al. (2013).

# 7.1 Evaluation factor 1 – Feasibility of making a post-closure safety assessment

This evaluation factor can be divided in two parts:

- A) Process understanding. The level of process understanding underpins the treatment of the processes in the assessment; a firm understanding will enable modelling of the process in the safety assessment.
- B) The initial state. The initial state is the starting point for any safety assessment, and it is important that the initial state can be defined and achieved. Thus, the ease of defining and achieving the initial state needs to be considered.

Below, the different concepts are reviewed in light of this evaluation factor, and the different safety functions connected to the repository concepts.

#### Gravel repository

Some examples of the processes that affect the safety functions attributed to the gravel, high permeability, are:

- Clogging.
- Freezing.
- Water transport/Two-phase flow/Mixing.
- Dissolution/Precipitation.
- Colloid formation and transport.
- Microbial processes.

As for concrete, this is a widely used material for various constructions and purposes. Many of the relevant processes are well understood, theoretically, but there is not enough information available from relevant, long-term experiments for the purposes of modelling the change of hydraulic conductivity over time. If no significant changes occur in the hydraulic properties over time, the modelling should be relatively straightforward (Neretnieks and Moreno 2013). It is estimated that for a future safety assessment, reactive transport modelling would be required, which in turn is dependent on site specific data. Also, some model development would be necessary. With regards to initial state, no specific difficulties have been identified. Therefore, it is judged that the effort involved in making a safety assessment is likely less than for both the concrete and bentonite concepts. Lack of long-term studies of clogging adds to the uncertainty, as does the lack of specific site data.

#### Concrete repository

The processes that affect the safety functions attributed to the concrete, namely low groundwater flow, low diffusivity, strong sorption, and high-pH conditions, are, among others:

- Water transport.
- Freezing.
- Fracturing.
- Dissolution/Precipitation.
- Speciation and sorption.
- Colloid formation and transport.

All of these are important to understand when considering concrete degradation in general. There is a difference in the level of understanding when it comes to pristine and aged concrete. It is here judged that the understanding of, for example, water transport, speciation and sorption in pristine concrete is such that radionuclide transport through the materials can be modelled (Neretnieks and Moreno 2013). The knowledge about properties of, and processes resulting in, degraded concrete, are less advanced. There is an ongoing and planned research program concerning degraded concrete (see Chapter 10 of Elfwing et al. 2013). With regards to initial state, it need to be emphasised that concrete is a widely used material and it is likely to be relatively straightforward to define the initial state. The task of achieving the initial state, as well as verifying it, might however pose some problems. This is recognised as an area where some technical development will be needed.

Importantly, even though further research is required concerning the degradation process, it is clear that pH in the pore water will remain high during the degradation. This is important especially for evaluating the effect of the concrete environment on the dissolution rate of metallic waste over time. The main uncertainties lie in the verification of the initial state, and the degradation processes.

#### **Clay repository**

The safety functions attributed to the bentonite are low groundwater flow, strong sorption, and low diffusivity, as well as filtering of colloids. The following list provides an overview of the many relevant processes connected to these functions that will be important to understand and to model:

- Water uptake and transport for unsaturated conditions.
- Water transport for saturated conditions.
- Piping/erosion.
- Swelling/mass redistribution.
- Liquefaction.
- Freezing.
- Water transport/Two-phase flow/Mixing.
- Dissolution/Precipitation.
- Speciation and sorption, including ion exchange.
- Diffusion/ Diffusive transport of species.
- Colloid release / Bentonite erosion.
- Colloid formation and transport.

All of these bentonite processes have been subjected to much research (SKB 2010c), and process understanding is advanced for this material. There is also substantial experience available for modelling of these processes, see for example Åkesson et al. (2010). Some areas will likely require further research, however. The effect of inhomogenous water saturation is one example. The proposed design of the Clay repository involves a very large amount of bentonite clay. The swelling during the saturation will need to be fully understood and described in order to evaluate the effects on the overall long-term permeability of the bentonite. Also, cement-bentonite interaction is identified as an important area which requires further research (Neretnieks and Moreno 2013). One process which may prove especially important for the SFL repository is the colloid transport through bentonite. This depends somewhat on the waste inventory and chemical properties of the waste; the legacy waste is complex and it is possible that some radionuclides may be transported through sorption on colloids which form as the waste degrades. In addition, some actinides, which are present in the legacy waste, can potentially form true colloids. The existing knowledge concerning the transport of colloids through bentonite suggests that it will be important to evaluate how well these different types of colloids will be filtered by the bentonite. For initial state verification, it will be essential to determine that the initial state is in fact achieved in the full volume of bentonite. Based on the above, there are identified uncertainties related to homogenisation processes (i.e. bentonite swelling model) and verification of the initial state, that may affect the ease with which one can perform the safety assessment for the Clay repository.

#### Super silo

The safety functions of the Super silo are based on the properties of the three barrier materials concrete, bentonite and gravel. Low groundwater flow, low diffusivity, and strong sorption are connected to both concrete and bentonite. The gravel around the silo is expected to function as a hydraulic cage. The idea is to, by combining barriers, strengthen the overall safety function of the repository, but it is recognised that the combination of materials also adds uncertainties and complexity. Interactions between the different barrier materials need to be understood and taken into account in the models. With regards to process understanding, the Super silo requires both further research and model development, mainly for bentonite-concrete interaction (Neretnieks and Moreno 2013). The envisaged method of waste and barrier emplacement also involves some important uncertainties related to the initial state, which needs attention. This relates, partly, to the effect on already installed barriers from processes occurring during the operational phase of the repository (see Grahm et al. 2013). In summary, the Super silo is judged to require substantial modelling effort and research, especially regarding physical and chemical interaction between bentonite and concrete.

# 7.2 Evaluation factor 2 – Robustness of the barrier safety functions

This evaluation factor relate to many different aspects of the proposed barrier system. The focus lies on the potential of the engineered barriers to provide and maintain the required hydrological, mechanical and chemical properties. The possible effects of processes in both waste and other barriers on a safety function should be evaluated, as should the durability of the barriers.

#### Gravel repository

The Gravel repository exhibits one safety function, namely that of the high hydraulic conductivity of the gravel. The most relevant processes that may affect this safety function are clogging, dissolution and precipitation, all of which will change the hydraulic properties of the gravel.

Even if it were to be shown that the hydraulic properties of the gravel fill may be maintained for a long time, a preliminary evaluation based on a modelling exercise (Neretnieks and Moreno 2013) indicates that the safety function high hydraulic conductivity does not contribute to retardation. Thus, it appears as if using only gravel as a barrier does not provide any effective safety functions. In the simple model, the waste is not protected by any means, and radionuclides will escape mainly by diffusion, but also partly by flow, from the waste to the flowing water.

It is still possible that if additional protective features are considered, or if the hydraulic cage concept is used in combination with other barriers, it may still contribute to safety by diverting the ground-water flow. Even so, there are still large uncertainties regarding how long the hydraulic cage will function as envisaged, and thus how long the safety function will be maintained. Clogging is likely to affect the hydraulic properties (e.g. Sternö 2005), and the performance of the hydraulic cage is likely very site specific. The long-term effect of dissolution and precipitation, processes expected in

the gravel, are uncertain. Another possible problem might be that high groundwater flow might cause the concentration at the waste-gravel interface to be very low, potentially increasing the diffusion rate of radionuclides from the waste to the to the high flow cage.

In summary, this preliminary evaluation of the Gravel repository indicates that the safety function connected to the gravel does not contribute to retardation. The Gravel repository concept could be improved by combining the hydraulic cage with additional barriers and/or features; however, there are still uncertainties relating to the negative effects of the high groundwater flow, and the possibility to maintain hydraulic conductivity for very long times.

#### Concrete repository

In a repository, concrete can be expected to degrade over time. Concrete degradation is expected to affect all safety functions: low groundwater flow, low diffusivity, strong sorption, and high-pH conditions. Initially, however, the concrete will be pristine, and amenable to an assessment of the proposed safety functions. This has been done, through preliminary modelling, by Neretnieks and Moreno (2013). The results of this modelling exercise show that a pristine concrete monolith should show excellent performance with regards to low hydraulic conductivity, slow diffusion and high sorption.

To assess the long-term performance of the concrete it is required to first identify the processes affecting the safety functions. Concrete degradation, which is expected to occur over time, involves fracture development, dissolution and precipitation. The chemical reactions that are connected to these processes change the chemical properties of the pore water, including pH. The reactions also affect many other properties of the concrete, and may initiate fracturing. Changes in the waste packages (e.g. expansion), or rock movements, can also cause fracturing. Dissolution and precipitation can cause changes in chemical properties affecting sorption, as well as hydraulic properties.

In general, the robustness of the safety functions depends on the rate of concrete degradation. However, the safety functions will be maintained for some time even when degradation processes are active. For example, it is expected that much of the sorptive properties will be maintained even though the mineralogical composition is changing. The alteration of concrete will also cause a slow change of the pH of the pore water; as long as pH is kept above a certain threshold, the passivation of metals is still expected to be functional. Considering the vast amounts of concrete which will be available, the pH can be expected to be buffered for a long period of time, providing a high-pH environment. This should be a robust safety function for the steel-based waste. It should be noted, however, that the leaching of the alkali and earth-alkali hydroxides of the concrete will also affect the surrounding chemical environment in the natural barrier, i.e. the hydrogeochemical properties of the rock system. There will be a high-pH plume in the groundwater which has interacted with the concrete.

As indicated in Neretnieks and Moreno (2013), the barrier safety functions of the Concrete repository are expected to initially be very strong. Degradation and fracturing are expected to affect the concrete over time, and there are uncertainties as at what point the safety functions are not effective any more. It is estimated that further research is required in order to evaluate how the processes related to degradation affect water flow, diffusion and sorption. The large amounts of concrete are expected to be beneficial for the robustness of the safety functions connected to chemical properties; pH buffering and sorption. The highly alkaline environment is expected to be maintained for a substantial amount of time after closure. The properties affecting sorption of different radionuclides are expected to change; however, the change may in some cases cause improved sorption. Thus sorption is potentially a robust safety function. In summary, initially all safety functions are expected to be strong, but over time the most robust safety functions may be the alkaline environment, as well as sorption (especially for some radionuclides). The long-lived alkaline environment is a significant consideration for the metallic waste.

#### Clay repository

The Clay repository contains vast amounts of bentonite. This is expected to ensure low groundwater flow, low diffusivity, and strong sorption. An additional and important safety function is the filtering of colloids. As long as the bentonite is in place and intact, these safety functions are expected to be effective. The robustness of safety functions connected to bentonite in a KBS-3 repository has recently been thoroughly evaluated (SKB 2011).

Compared with pristine concrete, the initial properties of bentonite is less effective for ensuring radionuclide transport by diffusion only (Neretnieks and Moreno 2013). This relates to the initial hydraulic conductivity of the pristine materials; this is higher for bentonite, and the retardation (from diffusion and sorption) is not as strong as in concrete (Neretnieks and Moreno 2013). For the evaluation performed here, however, the robustness of the barrier system needs to be considered, which means to take into account the potential degradation of the properties which provide the safety functions.

For bentonite, in a KBS-3 repository, the main threat appears to be diluted groundwater, which may cause bentonite erosion (see for example SKB 2011, p 529). However, in the Clay repository concept, very large amounts of bentonite are available to, through self-sealing, fill in any voids that may result from erosion by diluted water. A process which might affect the bentonite negatively over time is the interaction between bentonite and concrete. Concrete will be available in structural components in the core of the vault, and in the legacy waste, if this is not re-conditioned (Neretnieks and Moreno 2013). The bentonite-concrete interaction is expected to affect properties related to radionuclide sorption and diffusion through the bentonite; however, again the large amounts of bentonite may counteract and minimize the negative effects. This needs to be investigated further. Finally, the bentonite has the added function of effectively filtering colloids; this has bearing on the possible colloid facilitated transport of actinides, a possible problem mostly connected with the legacy waste. This safety function is expected to be maintained as long as the bentonite is more or less intact. To summarize, the bentonite in the Clay repository is expected to be robust, and safety functions maintained, for a long period of time.

#### Super Silo

The idea with the Super silo is to combine the safety functions of the concrete, bentonite and gravel to enhance the radionuclide retardation. Thus, it will here be evaluated how well the low hydraulic conductivity, low diffusivity, and strong sorption in both the concrete and the bentonite is likely to be maintained. The additional safety functions of bentonite and concrete, namely that of colloid filtering of bentonite, and high pH from the concrete, should also be evaluated. The gravel surrounding the silo is expected to function as a hydraulic cage, with high hydraulic conductivity. Using these multiple engineered barriers should reinforce the retardation of radionuclides; however, there are several uncertainties and issues that need to be addressed in the concept as it is defined.

The durability of the concrete barrier may be affected by the swelling bentonite, and the bentonite will be negatively affected by concrete. In a design involving a double concrete wall, it will be expected that the bentonite will be attacked from two sides, and lose the safety functions at the rate of the alteration caused by the bentonite-concrete interaction. The outer concrete wall may fracture and thereby the radionuclides will be transported by advective flow in the fractures away from the repository (Neretnieks and Moreno 2013). The situation might be improved if the design instead involves bentonite outside of a single concrete wall. The damaging effects of the bentonite-concrete interaction may be minimised by using low-pH cement; however, in order to evaluate the effects on the safety functions, further research efforts and model development are required.

One conclusion of the evaluation is, therefore, that the potential for the safety functions of the Super silo to remain effective for long periods of time depends partly on the design. The proposed design, with double concrete walls, is judged to be connected to a risk of severely damaging both the bentonite and the concrete walls. The Super silo still has potential, however, in the case of a modified design with bentonite placed outside of concrete. In that case, and with intact barriers, the retention of radionuclide is very effective; however, this is dependent on the effect of the bentonite-concrete interaction.

### 7.3 Summary of evaluation according to evaluation factors

Summarizing the results of the evaluations presented above, the first observation is that the Gravel repository is not promising, in terms of long-term safety. It is possible, that beneficial effects can still be attributed if the hydraulic cage concept is combined with some other feature or barrier that may improve radionuclide retention. The Concrete repository is promising with regards to the very good retention displayed by the intact, non-degraded concrete. It is expected that even a degraded concrete would contribute with high pH and strong sorption, thus slowing down the release of radionuclides, especially from metallic waste. The safety functions of the Clay repository are expected to be very robust, and the process understanding and modelling of the evolution of the properties of bentonite is advanced. Notable is also the colloid-filtering capacity of bentonite, which has the potential of slowing down transport of actinide colloids. Thus, also the Clay repository concept is promising. The Super silo combined different barrier materials, but even if the intact barrier system potentially has the strongest retention properties, the complexity of the design and the negative effects of bentonite-concrete interaction indicate that the safety functions may be lost prematurely. In addition, problems are identified when it comes to defining and achieving the initial state. If the design is changed and the knowledge concerning the effect of cement-concrete interaction is enhanced, it would improve the Super silo concept.

The preliminary evaluation presented here is summarized in Table 7-1. The most promising concepts, considering both evaluation factors, are the Concrete and Clay repository concepts. The Super silo, which combines three barrier materials, involves a number of uncertainties and potential problems, which affects the evaluation negatively in spite of the potential of the intact barrier structure in an ideal situation. The Gravel repository requires significant improvements, for example by adding barrier components.

Table 7-1. Overview of the evaluation of the concepts according to the Evaluation Factors.
Green: Promising, Yellow: Potential problem identified, Red: Problem(s) identified. The argu-
ments behind these evaluations are given in the text.

Evaluation factor	Gravel	Concrete	Clay	Super silo
Feasibility of making a post-closure safety assessment				
Process understanding				
Defining and achieving the initial state				
Robustness of the barrier safety functions				
Initial Hydraulic conductivity		_		
Initial Diffusivity				
Initial Sorption				
Other chemical properties (pH, colloid filtering)				
Long-term effects of waste processes				_
Long-term effects of barrier processes				
Barrier-barrier interaction (including natural barrier)				

### 8 Outlook and concluding remarks

The aim of this report is to give an account of progress regarding the evaluation of long-term safety of different repository concepts suggested for the SFL Concept study. Issues concerning long-term safety of a future SFL have been identified, and further investigations and analysis have been suggested to aid in the continued process of concept development. Four different concepts are described and subjected to a preliminary evaluation: the Gravel repository, the Clay repository, the Concrete repository and the Super silo. In addition, four different conditioning methods are described: Sealed container, Melting of metallic waste, Vitrification, and Chemically improved retention. The processes that are expected to affect the safety functions of the different barriers are described, as are processes in the natural system expected to be of importance in a future safety assessment. A comparison of the different concepts is presented, based on preliminary models of near-field transport capacity. Finally, an evaluation of the concepts is reported, taking into account the preliminary modelling and understanding of relevant processes.

#### Conclusions of the evaluations

The evaluation presented here shows that the most promising SFL repository concepts for further assessment are:

- The Concrete repository.
- The Clay repository.

Less promising, due to enhanced complexity and need to redesign, is:

• The Super silo.

Not promising:

• The Gravel repository.

In particular, it should be noted that:

- The Gravel repository will not function as a stand-alone concept; additional features or barriers are needed to improve the concept.
- Bentonite has potential to reduce colloid-facilitated transport of actinides, and is suggested as a suitable barrier for the legacy waste.
- Concrete is a suitable barrier material for the steel waste, as concrete will elevate pH and thereby lower the dissolution rate of that waste.

#### Possible combinations, improvements and alternatives

The two main types of long-lived waste intended for disposal in SFL, the legacy waste and the steelbased reactor components, are of very different character. One main, yet undetermined, factor that may affect the repository design and choice of barrier material, is the conditioning of the waste. The steel components are, today, more easily characterised, and more homogeneous compared with the legacy waste. The possibility of melting this waste, in order to lower the release rate, is interesting. It does, however, require a certain period of time for the activity to decay to levels that are acceptable for this method. If melting is utilized for this waste category, a re-evaluation of the concepts might be necessary. The same reasoning can be applied for the sealed container, if it can be shown that the weld of the sealed container will take as long time as the rest of the container to corrode. The current preliminary evaluation shows that the Gravel repository will not function as envisaged using an unprotected waste form. If, however, this concept is improved by using a waste form with a safety function which slows down the radionuclide release from the waste to the engineered barrier, this could warrant a new evaluation. This could be achieved with a sealed container, or drastically diminishing the surface area, and therefore the dissolution rate, of the metallic waste through melting and moulding. The legacy waste is very inhomogeneous, and difficult to characterise. It might be required to re-condition this waste before it can be shown to agree with acceptance criteria. Whatever the waste form or waste forms that are then produced might raise a need to reconsider the barrier alternatives. Of the possible additional features, that might improve retention, one does not specifically deal with the waste. The option to include a material which chemically improves the retention (through strong sorption), especially of anionic species, needs to be further evaluated. Of these types of materials Red Mud seems most versatile and promising, and research is ongoing internationally concerning the possibilities to combine Red Mud and concrete (e.g. Oliveira and Rossi 2012).

The weaknesses of the Super silo, considering the long-term safety, are focused around the bentonite-concrete interaction, but some real problems related to the initial state are also identified. This might be resolved through enhanced efforts in research and technology development, as well as a change in the design. The alternative to apply the silo geometry to the Concrete repository could be interesting, if it is shown that such geometry makes it easier to avoid intersecting fracture zones in the bedrock.

#### Suggested further research

In the further work planned in the framework of developing the SFL repository concept, or concepts, preliminary safety evaluations need to be carried out and their results need to be taken into account. This involves research relating to the whole system: the source term, the engineered barrier system, and the natural system.

Overall needs for the future safety assessments are investigations concerning:

- methods of how to calculate dose, and potential use of different types of calculations than previously,
- improvement of the connection between the different models describing the repository, the geosphere, and the biosphere,
- establishment of a synchronised landscape and climate development.

Areas for further research which mainly concern the different barrier materials, and processes affecting them:

- concrete degradation: effects on material strength, in hydraulic conductivity, and chemical properties,
- · concrete reinforcement: investigations of alternative reinforcement methods,
- gas formation and transport: This is relevant for both concrete and bentonite, and relates strongly to the waste form,
- bentonite homogenisation: bentonite swelling model for the Clay repository,
- bentonite-concrete interaction: significance for Clay repository, beneficial effects of low-pH cement.

If a future decision on the repository design involves gravel as a hydraulic cage around a barrier, the following is suggested:

- · detailed study of the water flow through the repository and radionuclide release rates,
- research relating to how the hydraulic properties of the gravel will change over time,
- study of the chemical evolution of the mineral surfaces of the gravel.

The results of the preliminary assessment will indicate what parts of the proposed repository design are most in need of improvement, as well as guide other parts of the process, for example the site selection.

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#### **Unpublished documents**

SKBdoc id, version	Title	lssuer, year
1416968 ver 1.0	Low and intermediate level waste in SFL3–5: Reference inventory	SKB, 1998