TECHNICAL REPORT

95-03

Prestudy of final disposal of long-lived low and intermediate level waste

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January 1995

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PRESTUDY OF FINAL DISPOSAL OF LONG-LIVED LOW AND INTERMEDIATE LEVEL WASTE

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Kemakta Konsult AB, Stockholm, Sweden

January 1995

This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

Information on SKB technical reports from1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17), 1982 (TR 82-28), 1983 (TR 83-77), 1984 (TR 85-01), 1985 (TR 85-20), 1986 (TR 86-31), 1987 (TR 87-33), 1988 (TR 88-32), 1989 (TR 89-40), 1990 (TR 90-46), 1991 (TR 91-64), 1992 (TR 92-46) and 1993 (TR 93-34) is available through SKB.

Prestudy of Final Disposal of Long-Lived Low and Intermediate Level Waste

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ABSTRACT

The repository for long-lived low and intermediate level waste, SFL 3-5, is foreseen to be located adjacent to the deep repository for spent encapsulated fuel, SFL 2. The SFL 3-5 repository comprises of three repository parts which will be used for the different categories of waste. In this report the work performed within a prestudy of the SFL 3-5 repository concept is summarised. The aim was to make a first preliminary and simplified assessment of the near-field as a barrier to radionuclide dispersion.

A major task has been to compile information on the waste foreseen to be disposed of in SFL 3-5. The waste comprises of; low and intermediate level waste from Studsvik, operational waste from the central interim storage for spent fuel, CLAB, and the encapsulation plant, decommissioning waste from these facilities, and core components and internal parts from the reactors. The total waste volume has been estimated to about 25 000 m^3. The total activity content at repository closure is estimated to be about $1 \cdot 10^{17}$ Bq in SFL 3-5. At repository closure the short-lived radionuclides, for example Co-60 and Fe-55, have decayed considerably and the activity is dominated by nickel isotopes in the metallic waste from the reactors, to be disposed of in SFL 5. However, other radionuclides may be more or equally important from a safety point of view, e.g cesium-isotopes and actinides which are found in largest amounts in the SFL 3 waste.

A first evaluation of the long term performance of the SFL 3-5 repository has been made. A systematic methodology for scenario formulation was tested. It was possible to carry through the different steps in the methodology, to formulate a Reference Scenario for SFL 3-5 and to define a Reference Case to be quantitatively analysed. An important result from this part of study is the documentation. The documentation comprises of the Influence diagram with the linked data base with documents, protocols and decisions behind the development of the Reference Scenario. This will facilitate future re-evaluations.

During the prestudy, investigations and experimental works have started up at different organisations to support the need for data in the evaluations of the SFL 3-5 repository performance.

There is a number of processes and mechanisms that may be of potential importance in the evaluation of long-term repository performance. In the prestudy, only simplified calculations have been performed on average hydraulic conditions in the repository, potential increase in temperature and potential gas formation in the waste packaging. The chemical conditions in the repository and factors influencing availability and retardation of elements have been considered in the selection of data for the Reference Case calculations.

The near-field release of contaminants was calculated for a selected number of radionuclides and chemotoxic elements. The radionuclide release calculations revealed that Cs-137 and Ni-63 would dominate the annual release from all repository parts during the first 1000 years after repository closure and that Ni-59 would dominate at longer times. The highest release rates arises from SFL 4, despite the fact that the total content of radionuclides are more than one order of magnitude less compared to the other repository parts. The main reason is that in the calculations retention of radionuclides is

only considered for the sand backfill in SFL 4. In order to get a measure of the radiotoxicity, the near-field releases were converted to intermediate doses by assuming that the entire release from the repository was captured in a drinking-water well. The intermediate doses were for all the studied radionuclides below 0.1 mSv/year for the simplified well scenario. Lead and beryllium were chosen as model substances for chemotoxic elements and included as an example in the release calculations for SFL 3. For the same simplified well scenario as for the radionuclide releases, it was found that the near-field releases of lead and beryllium would result in concentrations in the water in the well which are below the guideline values used for drinking water.

Finally, it must be remembered that the near-field release calculations have been performed on a preliminary waste inventory and for subset of elements, and the defined Reference Case does not consider all processes and mechanisms assessed to be of potential importance in the Reference Scenario. The importance of these processes and mechanisms have to be evaluated in future performance assessments. In addition, other scenarios for example describing the potential influence of initial conditions in the repository and changes of environmental conditions have to be studied.

SAMMANFATTNING

Ett förvar för långlivat låg- och medelaktivt avfall, SFL 3-5, planeras att samlokaliseras med djupförvaret för använt kärnbränsle, SFL 2. I denna rapport ges en översikt av det arbete som utförts inom en förstudie av SFL 3-5. Målsättningen med studien har varit att göra en första bedömning av förvaret och närområdet som barriärer mot utsläpp av radionuklider och kemotoxiska element.

Inventering av avfall som kan komma att deponeras i SFL 3-5 har varit en huvuduppgift. Avfallet består av låg och medelaktivt avfall från Studsvik, drifts- och rivningsavfall från mellanlagret för använt kärnbränsle och inkapslingsstationen, samt härdkomponenter och interna delar från reaktorerna. Mängden avfall till SFL 3-5 har uppskattats till ca 25 000 m^3. Den totala aktiviteten i SFL 3-5 vid förslutning, år 2040, har uppskattats till ca $1 \cdot 10^{17}$ Bq. Vid den tidpunkt då förvaret försluts domineras aktivitetsinnehållet av nickelisotoperna i det metalliska avfallet från reaktorerna, som skall deponeras i SFL 5. Då har kobolt, som dominerar aktiviteten i avfallet initialt, klingat av avsevärt. I SFL 3 återfinnes de största mängderna av cesiumisotoper och aktinider vilka kan vara minst lika betydelsefulla som nickel ur säkerhetssynpunkt.

I utvärderingen av den långsiktiga säkerheten av ett förvar spelar valet av scenarier en avgörande roll. Inom förstudien har en systematisk metod baserad på Influensdiagram provats för att formulera ett Referensscenario för SFL 3-5 och för att definiera ett Referensfall för kvantitativ analys. Ett viktigt delresultat från denna övning är den dokumentation som sammanställts. Dokumentationen består av beskrivningar av komponenterna i Influensdiagrammet samt protokoll över de val och beslut som ligger till grund för framtagandet av Referensscenariot. Denna information kan förenkla framtida utvärderingar.

Under förstudien har utredningar och experiment vid olika organisationer initierats för att stödja behovet av data i den framtida utvärderingen av förvarets långsiktiga funktion.

Vid analysen av utsläppet av radionuklider och kemotoxiska element från ett förvar finns det många processer och mekanismer som måste beaktas för att utvärdera förvarets långsiktiga funktion. I förstudien har förenklade beräkningar genomförts avseende vattenflödet i förvaret, temperaturökning och gasbildning. I utsläppsberäkningarna har tillgängligheten och fördröjningen av elementen samt de kemiska förhållandena i förvaret beaktats i valet av data för Referensfallet.

Utsläppet från närzonen har beräknats för ett begränsat antal radionuklider och kemotoxiska element för Referensfallet. Under de första 1000 åren efter förslutning domineras det årliga utsläppet av Cs-137 och Ni-63, och vid längre tider av Ni-59. Det högsta utsläppet kommer från SFL 4 trots att den har betydligt lägre aktivitetsinnehåll än övriga förvarsdelar. Detta kan förklaras av att endast fördröjning av radionuklider i sandåterfyllnaden beaktats i beräkningarna för SFL 4. Radionuklidutsläppet från närzonen har med hjälp av ett förenklat brunnsscenario omvandlats till dos. Närzonsdosen överstiger inte för någon av de studerade nukliderna 0.1 mSv/år. Bly och beryllium i SFL 3 har valts som exempel på utsläpp av kemotoxiska element. Med samma förenklade brunnsscenario som använts för att beräkna närzonsdoser resulterar närzonsutsläppet av

bly och beryllium i koncentrationer i brunnsvattnet som understiger gällande dricksvattennormer för dessa element.

Slutligen bör det påpekas att de närzonsutsläpp som redovisas för Referensfallet endast är att beakta som en indikation på förvarets funktion. Beräkningarna har genomförts på ett preliminärt avfallsinventar och för ett begränsat antal radionuklider. Det definierade Referensfallet inkluderar inte alla de processer och mekanismer som befunnits vara av potentiell betydelse för den långsiktiga säkerheten i Referensscenariot. Betydelsen av hittills ej beaktade processer och mekanismer samt eventuella förändringar i långtidsfunktionen för andra scenarios, t ex påverkan av initiala och förändrade yttre förhållanden, behöver studeras och utvärderas i framtida analyser.

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1 Introduction

1.1 Background

The Swedish deep repository for long-lived radioactive waste, SFL, is planned to be situated at a depth of about 500 m in the crystalline bedrock, see Figure 1.1. It consists of a repository intended for encapsulated spent fuel, SFL 2 and a repository intended for long-lived low and intermediate level waste, SFL 3-5. A previously planned repository for vitrified waste from reprocessing, SFL 1, has been omitted. A first outline of the design for SFL 3-5 was made in 1982 [*PLAN 82*] and a more detailed repository design is presented in [*PLAN 93*].

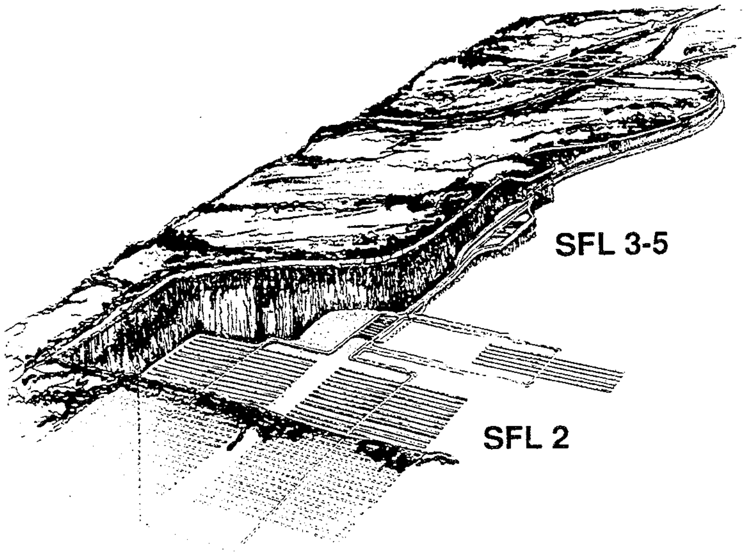

Figure 1.1 Artist's impression of the Swedish deep underground repository for long lived radioactive waste.

The SFL 3-5 repository comprises of three areas that will be used for the different categories of waste:

- SFL 3 for waste from Studsvik, the central interim storage for spent fuel, CLAB and the encapsulation plant, in operation
- SFL 4 for decommissioning waste
- SFL 5 for reactor components.

Strictly taken, not all of the waste intended for disposal in SFL 3-5 falls into the category of long-lived waste. In fact, only the waste that comes from Studsvik, the core components and the reactor internals are long-lived waste. Operational waste and later decommissioning waste from CLAB and the encapsulation plant could in principle be disposed of in the final repository for reactor waste, SFR. However, SFL 3-5 is intended to receive all low level waste, LLW and intermediate level waste, ILW, that arises in the post-closure period of SFR. That will include for example filter masses consisting of ion exchange resins which would also have been suitable for disposal in SFR.

The operation of SFL 3-5 and earliest time of closure and sealing are connected to the management and deep disposal of spent fuel. The reason for that is that SFL 3-5 will receive waste from spent fuel facilities such as CLAB and the encapsulation plant, including finally the decommissioning waste from these facilities. The emplacement of encapsulated spent fuel will be part of the first phase of operation of the deep repository which is expected to take place in the period 2009 to 2012. An evaluation will be made before further expansion. After that the remaining canisters are assumed to be deposited during the period 2020 to 2040. This second phase will presumably also include the operation of SFL 3-5. The repository parts SFL 3 and 5 will be needed for disposal operations before SFL 4 which consists of the repository tunnels. SFL 4 is intended for decommissioning waste and disused transport containers and is not needed until the final stage. According to this scheme the earliest time of closure and sealing of the repository area SFL 3-5 would be the year 2040. The probable procedure will be to start by closing SFL 3 and 5, and leave SFL 4 open for some additional years in order to receive decommissioning waste etc. However, in the prestudy we have decided to select 2040 as the reference time for all disposal parts, which implies that the individual estimated quantities of radionuclides will be set by their decay to the time 2040 before presented in tables in this report or the waste characterisation report [*Lindgren et al., 1994*]. This will also be the time of maximum radionuclide content in the repository SFL 3-5.

1.2 Aim of the study

At the end of 1992 [*SKB RD&D-PROGRAMME 92*] it was decided to begin the investigations concerning long-lived low and intermediate level waste than spent fuel by the following aims:

- Make an inventory of existing waste and make a forecast of produced waste intended for SFL 3-5.
- Continue work on the design of the repository for long-lived LLW and ILW.
- Prepare the safety assessment and gather data that will become necessary at a later stage.

In order to make an inventory and characterise the waste, it is not enough to consider just the radionuclide content. It is also important to know the material composition of the waste, how the waste have been conditioned and packed, etc.

Long-lived LLW and ILW are present in other countries too. Large quantities are obtained in countries that send spent fuel to reprocessing, for example France, the UK, Germany, Japan, USA and Switzerland. During reprocessing, long-lived radionuclides - mainly transuranic elements - are released from the fuel matrix when this is chemically dissolved. Most is reused, e.g. plutonium and uranium, or ends up in the high-level waste, but some long-lived radionuclides end up in the LLW and ILW. Many of these countries have well advanced programs for planning, repository design and studies related to the safety performance. An important task for SKB is therefore to follow the developments in other countries. In an international comparison, our quantities of this waste are small, since Sweden does not intend to reprocess the spent fuel.

An informal exchange of experience have been established between SKB and the organisations ANDRA (France), NAGRA (Switzerland) and NIREX (the UK). This is reflected in this study, where recently published data from NAGRA and NIREX have been particularly useful.

However, just gathering information on the properties of Swedish long-lived waste and following the international development in the field of performance assessment is not enough to achieve the aims set up in the RD&D-Programme. It will be necessary to perform experimental investigations which are not carried out elsewhere and there are models that need to be tested and adopted to the conditions specific for our disposal concept. Some of the experiments take a long time to carry out and need to be initiated in time, and it is important to know which experiments are relevant and really needed. It was therefore decided to perform a preinvestigation, starting at the beginning of 1993 and intended to end in spring 1994. The main goal of the preinvestigation was the following:

- To make a first preliminary and simplified assessment of the near-field barriers to radionuclide dispersion.

The prerequisites for this investigation was the conceptual design as it is presented in [*PLAN 93*] and general assumptions concerning the repository rock conditions. The study have been carried out in the form of a small project, which contained the following parts:

- An inventory and characterisation of the waste. Radionuclide content and other safety relevant components were summarised in an attempt to come as close as possible to the actual content of radionuclides, metals, organic materials, etc. Estimates have been made were hard data are not available yet.

- An inventory have been made of Features, Events and Processes (FEPs) that may influence the performance of the repository barriers to radionuclide release. Premises for a "Reference Scenario" were defined and the importance of identified FEPs for this "Reference Scenario" were evaluated. This formed the basis for the selection of a "Reference Case" which has been analyzed within the frame of the prestudy. This is a relatively new approach, and the exercise has contributed to the development in this relatively new field which is frequently referred to as the scenario method for performance assessment.

- Laboratory experiments and literature studies of important chemical properties. This includes cellulose degradation, concrete leaching, radionuclide sorption and diffusion in concrete and bentonite.

- Selection of data and calculations of near-field release in the "Reference Case", which was taken from the scenario study.

To make a performance assessment, even in this simplified and preliminary form, is an effective way to focus the search for relevant data and direct the future investigations. This aspect has also been promoted by the close cooperation between experimentalists and modellers engaged in the prestudy project. Further, the waste characterisation in combination with performance assessment illustrates what needs to be checked when waste is being conditioned and packed. Experts engaged in this field have been participating in the project.

No attempt has been made to change the conceptual design given in [*PLAN 93*], and so far it has proven to be functional. A rough comparison has been made between the present design of SFL 3 and a silo alternative, but they turned out to be fairly equal. One design detail, that need to be better outlined is the SFL 4 which is a bit too vague in the present concept. The question whether the bentonite backfill really is an advantage in SFL 3 remains. A more comprehensive analysis of the "Reference Scenario" and additional scenarios has to be made before an answer can be obtained, and that was not possible to achieve within the prestudy.

1.3 Presentation of working group and procedure

The prestudy project of waste disposal safety in SFL 3-5 was carried out by a group of specialists meeting at regular intervals of about once per month. The main fields covered by the working group were repository performance assessment, radioactive waste management and chemistry of radionuclides and repository environments. Less emphasis was put on other issues that need to be covered for a complete safety assessment, for example hydrogeology. However, that and related geoscientific studies will become increasingly more important as we come closer to the selection of a repository site. Within the frame of the prestudy it was possible to use generic information from related investigations. We have simply used the results from the studies made for SFR in Forsmark and for the spent fuel repository SFL 2.

The following persons participate in the prestudy and have been active in the writing of this report:

Albinsson Yngve	Chalmers University
Allard Bert	University of Linköping
Brodén Karin	Studsvik RadWaste AB
Carlsson Jan	SKB
Eng Torsten	SKB
Engkvist Ingmar	Chalmers University
Johansson Marie	SKB
Karlsson Fred	SKB (Chairman)
Lindgren Maria	Kemakta Konsult AB
Nordlund Eva	University of Linköping
Skagius Kristina	Kemakta Konsult AB
Wiborgh Marie	Kemakta Konsult AB

The prestudy project was managed by SKB and this report has been edited by Marie Wiborgh.

1.4 Initiated activities within the study

During the prestudy in 1993, and to some extent as a result of the fact that data needs to be developed in the course of the performance assessment, the following investigations were initiated at different organisations:

- Batch studies of radionuclide sorption. Crushed concrete from SFR is used as the solid substrate. Both normal construction concrete and porous backfill concrete is represented. Fresh cement porewater and leached concrete porewater are used as liquid phase. Radionuclides are used and the elements studied are: Eu, Th, Np, Am, Cm, Pm, Co, Ra, Ni and Cs. For some of the elements the dependence on concentration is being studied too. *Dept. of Nuclear Chemistry, Chalmers University of Technology.*

- Radionuclide diffusion in cement paste. The elements studied are: Ni, Cs and T (in the form of tritiated water). *Dept. of Nuclear Chemistry, Chalmers University of Technology.*

- Static leaching of cement paste. Synthetic groundwater is used to simulate deep geochemical conditions such as Allard water (normal composition) and NASK water (saline). The ions which are analyzed in the leachate are: Na^+, K^+, Ca^{2+}, Mg^{2+}, OH^-, NO_3^-, SO_4^{2-}, Cl^- and Si_{tot}. *Dept. of Nuclear Chemistry, Chalmers University of Technology.*

- Radionuclide diffusion in a 85/15 mixture of sand/bentonite (Wyoming MX-80) in a cement dominated chemical environment. Radionuclides are used and the elements studied are: Cs, Tc and Ni. *Dept. of Nuclear Chemistry, Chalmers University of Technology.*

- Solubility measurements of Ni, Pu and Eu under cementitious chemical conditions. *Dept. of Nuclear Chemistry, Chalmers University of Technology.*

- Studies of the formation of poly hydroxy compounds during the degradation of cellulose in an aerobic or anaerobic environment at pH 10 and 12. The rate of degradation is followed by UV-scanning, analysis of the total organic carbon (TOC) and identification of degradation products by the techniques Capillary Electrophoresis and GC/MS (Gas Chromatography coupled to Mass Spectrometry). *Dept. of Water and Environmental Studies, Linköping University.*

- Chemical degradation of gluconic acid in an aerobic or anaerobic environment at pH 10 and 12. The rate of degradation is followed with the same technique as previously described for cellulose degradation experiments. *Dept. of Water and Environmental Studies, Linköping University.*

- A library of model compounds have been compiled in order to identify the degradation products from cellulose and gluconic acid by capillary electrophoresis. *Dept. of Water and Environmental Studies, Linköping University.*

- The formation of metal complexes with poly hydroxy compounds is being investigated. *Dept. of Water and Environmental Studies, Linköping University.*

- The influence of poly hydroxy compounds on the absorption of metal ions on concrete and rock minerals is being studied. *Dept. of Water and Environmental Studies, Linköping University.*

- The present state of knowledge on concrete stability under deep repository conditions is being summarised. *Swedish Cement and Concrete Research Institute, Stockholm.*

Most of these investigations have continued into 1994. Results obtained so far have been used in the prestudy.

In addition to the above mentioned studies there are also investigations supported by adjacent research programmes at SKB that are expected to provide valuable insight and data for future performance assessments of SFL 3-5. Most promising are the study of the natural cement analogue in Jordan, field studies in connection with the SFR facility in Forsmark and the Äspö Hard Rock Laboratory near Oscarshamn in SE Sweden and laboratory studies of the interaction of cement porewater with granitic rock and bentonite clay minerals. Many of these studies are carried out in cooperation with organisations from other countries.

1.5 Structure of report

This report gives a summary of work performed within the prestudy for SFL 3-5. When reading the report it is essential to keep in mind that here is presented the first estimates on the near-field performance with supporting background information and not a complete performance assessment.

In the first chapters, the entities describing the studied repository system are presented. In chapter 2 an overview of the deep repository concept for long-lived waste, SFL, outlined in [*PLAN 93*] is described together with more detailed information on the SFL 3-5 repository concept. A summary of the waste inventory is presented in Chapter 3. The given estimates of amounts and composition of the waste intended for SFL 3-5 is a result from an investigation performed within the prestudy presented in the report "Low and Intermediate Level Waste for SFL 3-5" [*Lindgren et al., 1994*]. In Chapter 4 general barrier properties are presented for the foreseen barrier materials in the different repository parts. The repository is assumed to be located in general type Swedish bedrock and the repository site conditions are presented in Chapter 5.

In the selection of a "Reference Scenario", to form the base for the first estimates on near-field performance, a methodology based on the use of Influence diagrams with coupled documentation has been tested for SFL 3-5. The work is presented in the report "Testing of Influence Diagrams as a Tool for Scenario Development by Application on the SFL 3-5 Repository Concept" [*Skagius and Wiborgh, 1994*] and summarised in Chapter 6.

In the assessment of long-term repository performance the evolution of the conditions in the repository as well as processes potentially influencing the release of contaminants must be considered. In this report some of the topics are studied. In Chapter 7 the chemical conditions in the repository and factors influencing availability and retardation of elements are discussed. Estimates on the hydraulic and the thermal conditions in the repository are presented in Chapter 8. An overview of gas formation processes and estimates of gas formed by corrosion, microbial degradation and radiolysis are presented in Chapter 9.

The estimated near-field releases for the selected "Reference Case" defined by the "Reference Scenario" are presented in the report "Radionuclide release from the near-field of SFL 3-5. A Preliminary Study" [*Lindgren and Pers, 1994*] and summarised in Chapter 10.

Finally, in Chapter 11 the performed investigations within the prestudy are discussed and concluding remarks on the results are given in Chapter 12.

2 Description of repository concept

This study is based on the preliminary deep repository design presented in [*PLAN 93*]. The description of the SFL 3-5 repository concept has to some extent been detailed to support the performed estimates on near-field releases.

2.1 General overview of SFL repository

The deep repository for long-lived waste, SFL, is planned to be situated at about 500 m depth in the crystalline bedrock. SFL comprises of a deep repository for encapsulated spent fuel, SFL 2, and a deep repository for long-lived low and intermediate level waste, SFL 3-5. An overview of the present design of the deep repository for long-lived waste is shown in Figure 2.1.

SFL 3-5 is planned to be situated about 1 km away from SFL 2, and they are reached through the same, centrally located, shafts. Horizontal transport tunnels connect the two repository areas. The transport tunnels are foreseen to be sealed with a sand/bentonite mixture in the same way as the deposition tunnels in SFL 2. An alternative to shafts for the access from the surface is a ramp.

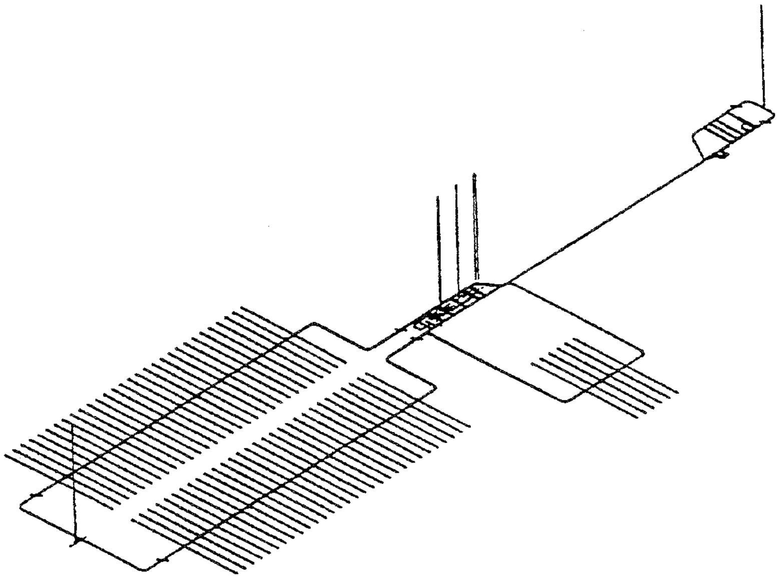

Figure 2.1 Overview of the SFL repository, SFL 2 is to the left and SFL 3-5 to the right.

A summary of foreseen wastes and main features of the SFL repository concept is presented below:

SFL 2	is intended for encapsulated spent fuel. The repository consists of a tunnel system where the canisters are embedded in bentonite and deposited in vertical holes in the tunnel floor.
SFL 3	is intended for long-lived or toxic waste from Studsvik, for example waste originating from research, industry and medicine. It shall also receive operational waste from the central interim storage for spent fuel, CLAB and the encapsulation plant, after the year 2010. In the repository vault a number of concrete compartments divided into a number of cells is foreseen to be constructed. The waste packages are to be stored in the concrete cells which are foreseen to be backfilled with porous concrete. The outside of the whole concrete structure will be surrounded by clay barriers.
SFL 4	is intended for decommissioning waste from CLAB and the encapsulation plant and for transport casks and containers. The repository comprises of the transport tunnels and the vaults left after sealing of SFL 3 and SFL 5. The waste packages are foreseen to be surrounded by a sand/gravel backfill only.
SFL 5	is intended for core components and internal parts from the nuclear power reactors. The repository vaults are foreseen to be divided into a number of concrete compartments. There are presently no plans to backfill the compartments but the outside of the concrete structures will be surrounded by a sand/gravel backfill.

2.2 SFL 2

SFL 2 is intended for encapsulated spent fuel. The spent fuel is encapsulated in steel canisters covered with a corrosion resistant layer of copper. A total of 4500 canisters will be deposited in SFL 2. In an early planning stage also a separate disposal part, SFL 1, for high level radioactive glass from reprocessing was discussed. Today this option has been withdrawn.

A schematic illustration of SFL 2 is shown in Figure 2.2. The facility consists of a system of horizontal disposal tunnels. The canisters will be disposed of in vertical holes, drilled in the bottom of the tunnels. The space between a canister and the rock will be filled with compacted bentonite clay. After disposal the tunnels are backfilled with a sand/bentonite mixture.

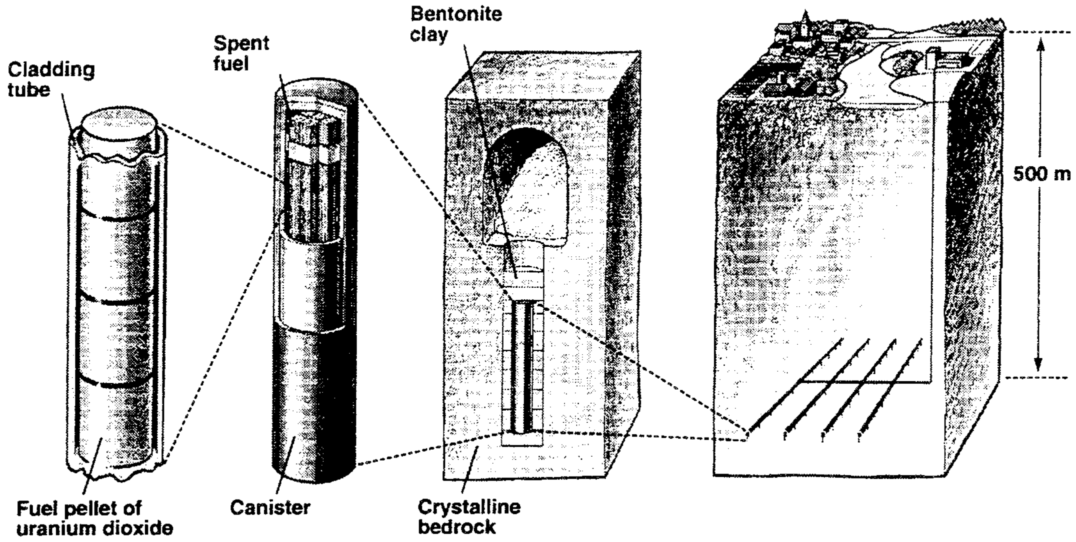

Figure 2.2 Schematic design of a deep repository for encapsulated spent fuel.

2.3 SFL 3-5

SFL 3-5 is intended for long-lived low and intermediate level waste. The total excavated rock volume for SFL 3-5 will be about 110 000 m^3 [*PLAN 93*] and the total volume of waste is estimated to about 25 000 m^3 [*Lindgren et al., 1994*]. An overview of the SFL 3-5 repository is shown in Figure 2.3.

The repository consists of three repository parts, SFL 3, SFL 4 and SFL 5. The repository parts are foreseen to host different kinds of waste types and have therefore different design and engineered barrier system.

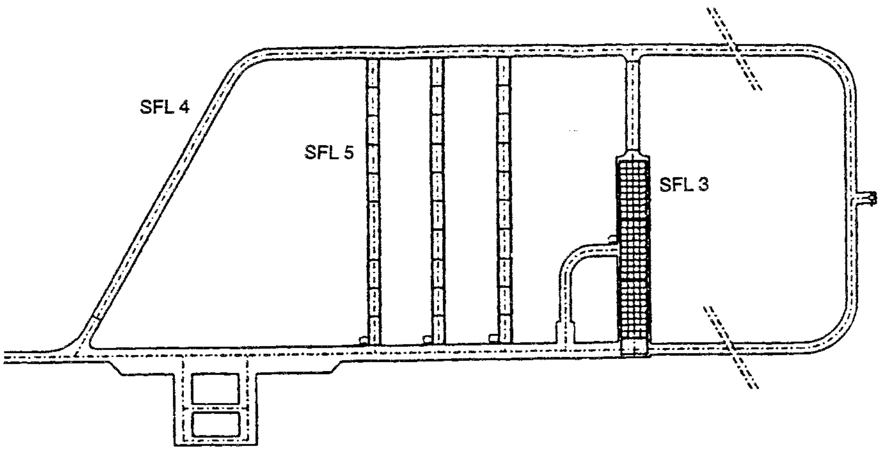

Figure 2.3 Overview of SFL 3-5.

2.3.1 SFL 3

SFL 3 is intended for long-lived or toxic waste from Studsvik and low- and intermediate level operational waste from CLAB and the encapsulation plant. The waste volume to be hosted in SFL 3 has been estimated to about 5000 m^3.

The waste from Studsvik consists both of waste from research at Studsvik and of waste collected from other users of radioactive materials in Sweden. The waste is mainly solid material placed in drums and in concrete or steel containers. Non-solid wastes (ion-exchange resins and sludges) are solidified in cement before disposal.

The operational waste from CLAB and the encapsulation plant consists of ion-exchange resins and filter aid materials conditioned with cement in concrete moulds and solid waste from service and maintenance placed in concrete moulds and backfilled with concrete.

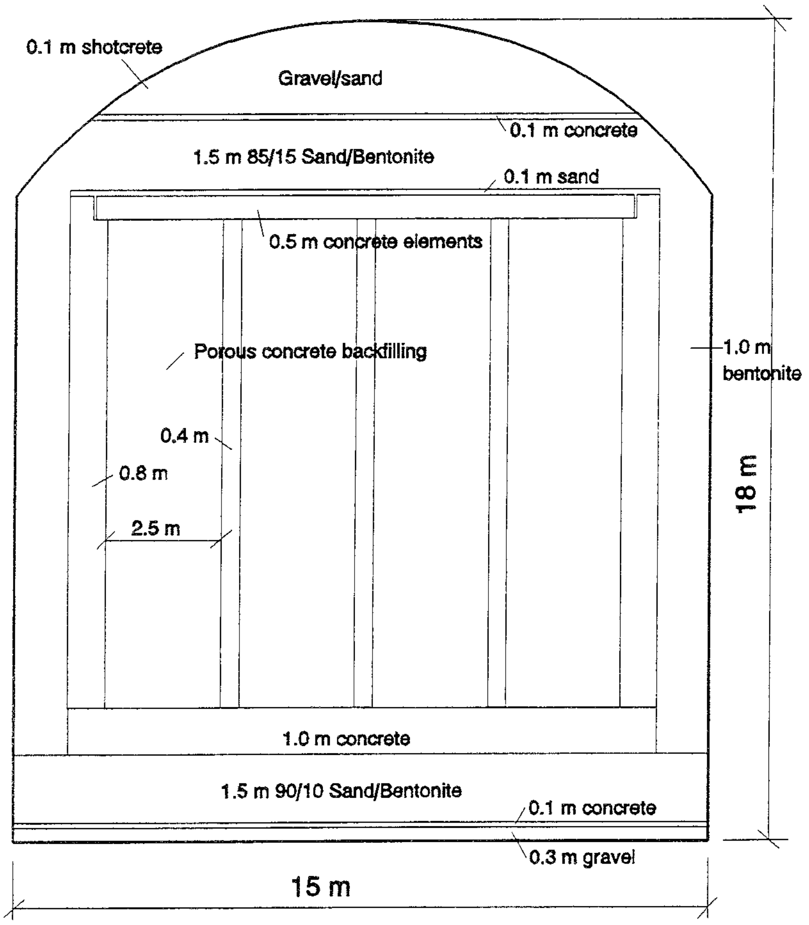

Figure 2.4 Schematic illustration of SFL 3.

In the present concept the SFL 3 vault is about 80 m long, 15 m wide and 18 m high, see Figure 2.4. The design of the interior of the vault is similar to the silo in SFR 1. The walls of the vault are covered with shotcrete. In the bottom of the vault a 0.1 m thick concrete plate will be casted above a 0.3 m thick drainage layer of gravel. Above the concrete plate a bed of 1.5 m 90/10 sand/bentonite mixture is placed on which a concrete structure is built. The waste containers/moulds are stacked in squared concrete cells, 2.5 ×2.5 m and the remaining empty space in the cells is filled with porous concrete. The space between the concrete cells and the rock, in average 1 m, is filled up with bentonite. Above the concrete structure a lid of concrete elements will be placed covered by a thin layer, about 0.1 m thick, of sand before the 1.5 m thick layer of 85/15 sand/bentonite is emplaced. Finally, above a supporting concrete plate, about 0.1 m thick, the remaining space will be filled with gravel or sand.

2.3.2 SFL 4

SFL 4 is intended for the fuel storage canisters from CLAB and the decommissioning waste from CLAB and the encapsulation plant as well as for transport casks and transport containers. The volume required by the waste in SFL 4 has been estimated to be about 10 000 m^3.

The decommissioning waste comprising of steel and concrete materials and the fuel storage canisters are foreseen to be emplaced in steel vessels without any backfill. The transport casks and containers are foreseen to be emplaced without any further packaging or shielding.

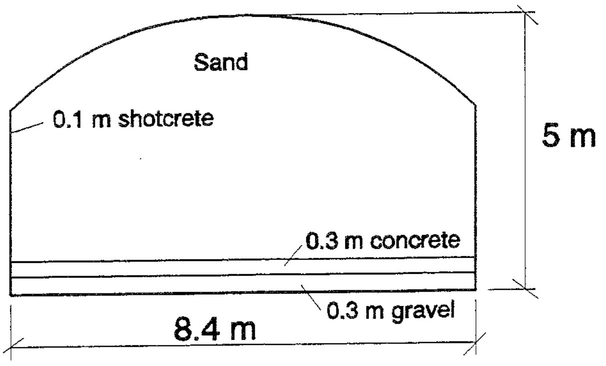

Figure 2.5 Schematic illustration of SFL 4.

SFL 4 is planned to consist of the tunnel system remaining when deposition in SFL 3 and SFL 5 has been concluded and the vaults sealed (see Figure 2.2). The total length of the tunnel system is about 500 m. The main part of the tunnel system is 8.4 m wide and 5 m high, see Figure 2.5. The walls are covered with shotcrete and in the bottom a 0.3 m thick concrete floor will be casted above a drainage layer of 0.3 m gravel. The waste will be placed in the tunnel system, which thereafter will be backfilled with sand.

2.3.3 SFL 5

SFL 5 is intended for core components and internal parts from the nuclear power reactors. The estimated waste volume is about 10 000 m^3 for the SFL 5 waste.

The waste is produced both during operation and decommissioning and comprises mainly steel components. The waste will be stored in steel cassettes which will be placed inside concrete containers and finally backfilled with concrete.

SFL 5 consists of three vaults, each 130 m long, 7 m wide and 10.5 m high. A schematic picture of the present design of SFL 5 is given in Figure 2.6. A concrete structure rests on top of a 0.3 m drainage layer of gravel. The structure, consisting of a bottom, outer walls and inner walls, makes compartments with space for 50 concrete containers each. The three vaults are foreseen to be divided in ten compartments each and can host 1500 concrete containers in total. After loading, a lid of concrete elements is placed on top of the compartments and the remaining void in the vaults is foreseen to be filled with sand.

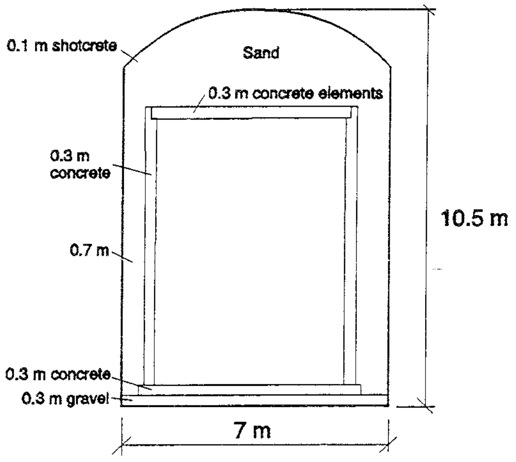

Figure 2.6 Schematic illustration of SFL 5.

2.4 Sealing of repository

With the major aim to prevent large quantities of water from flowing through and between repository parts, each repository part will be sealed.

After concluded deposition of waste and backfilling of the disposal vaults, each part will be sealed with plugs of "host rock quality" at the ends. The plugs consist of concrete, possibly in combination with compacted bentonite as sealing between the plug and the rock.

Finally, the entire tunnel system of the facility is planned to be backfilled with sand/bentonite (85/15-mixture). Before emplacement of the sand/bentonite in the tunnel between SFL 2 and SFL 3-5 the shotcrete on the walls as well as gravel and concrete on the tunnel floor is foreseen to be removed.

3 Waste description

Within the prestudy for SFL 3-5 a first inventory has been performed of the waste foreseen to be allocated to the repository [*Lindgren et al., 1994*]. Here a summary of waste origin, waste packaging, estimates of radionuclide content and potential content of toxic elements is presented for the individual repository parts SFL 3, 4 and 5. The presented waste inventory forms the base for the performed estimates on temperature, gas formation and contaminant releases from the near-field. However, the uncertainties in these estimates are large especially regarding old conditioned waste from Studsvik. Ongoing projects at Studsvik will however give more accurate information about the waste.

3.1 SFL 3 waste

General

The SFL 3 is designed for long-lived low level waste, LLW, and intermediate level waste, ILW, conditioned at Studsvik. Some of the radioactive waste from Studsvik does also contain toxic metals like for example lead, cadmium and beryllium. SFL 3 shall also receive operational waste from the central interim storage for spent fuel, CLAB and the encapsulation plant. In SFL 3 both organic and inorganic materials will be disposed of.

The waste from Studsvik consists of waste from research carried out at Studsvik and also of waste collected from other users of radioactive materials in Sweden e.g. industry, universities and hospitals. The raw waste materials are e.g. activated and contaminated scrap metals, precipitates, ashes, ion exchange resins, glove boxes, radiation sources, laboratory outfit and radiation protection equipments. Steel drums or concrete containers are mainly used as waste packagings.

Operational waste from CLAB and the encapsulation plant will be disposed of in SFR 1 as long as it is in operation. Waste produced thereafter will be disposed of in SFL 3. The waste consists of ion-exchange resins, filter aid and solid waste from service and maintenance. Concrete moulds with the same outer dimensions as the concrete containers for Studsvik waste are used as packagings.

Raw waste description and origin

The Studsvik waste to SFL 3 will include packages produced at Studsvik with raw waste originating from different facilities at Studsvik (Active Central Laboratory ACL, materials test laboratory α-Lab, Hot Cell Laboratory HCL, research reactor R2, fuel element storage FA and Van de Graaff-accelerator) and also from the national defence research establishment FOA, universities, hospitals, R1-reactor, the Army and industry. The raw waste consists for example of:

- ion exchange resins from R2 and FA (in most cases conditioned with concrete after treatment)

- liquid waste from ACL, HCL, R2 and universities (in most cases precipitations conditioned with concrete after treatment)
- exchanged activated and contaminated components from R2
- contaminated tools, instruments, filters, articles of consumption, laboratory outfit and radiation protection equipments from ACL, α-Lab, HCL, R2, FOA and universities
- plutonium contaminated glove boxes from ACL and FOA
- aluminium covered cadmium plates from R1
- radium night sights from the Army
- uranium waste from Asea-Atom (ashes after treatment)
- tritium in titaniumhydride from R2.

The packages may also contain short lived intermediate level raw waste that during storage in a concrete plug hole store at Studsvik has been cross contaminated by long lived waste, and short lived low level raw waste in the same packages as long lived waste. The intermediate level waste consists e.g. of radiation sources and exchanged components from different facilities at Studsvik. The low level waste consists e.g. of clothes, plastic bags, radiation sources, metal parts (different metals, different shapes) and construction materials from different facilities at Studsvik and other producers.

Operation of CLAB includes receiving of spent fuel and core components from the nuclear power plants, repackaging and storage in water filled pools. Radioactive waste consists mainly of crud from the fuel receiving operations for fuel (cooling) and ion exchange resins from the cleaning of the storage pool water. Minor quantities of metal scrap and trash from maintenance and replacement of equipments are generated.

The radionuclide inventory of the waste from CLAB is similar to that from the spent fuel storage pools at the nuclear power plants. This means that all waste from CLAB are suitable for disposal in SFR 1. However, as the operation of CLAB will continue for about 30 years after the closure of SFR 1, the waste production during that period has to be allocated to SFL 3 - although it is considered to be short lived waste.

The encapsulation plant will be in full-scale operation for 20 years from the year 2020. A shorter period of demonstration operation is anticipated before 2020 [*SKB RD&D-Programme 92*]. It is reasonable to assume that the waste from the demonstration operation will be treated with a delay of a few years after it has been generated. Therefore it can be appropriate to allocate all waste from the encapsulation plant to SFL 3.

It is anticipated that the encapsulation operations in the encapsulation plant and, later, the treatment of core components will give rise to approximately the same type and amount of waste as the receiving operations in CLAB. As very little is known about the handling of old spent fuel, this assumption will have to be reconsidered in the future.

Packaging and conditioning

Mainly reinforced cubical containers (outside dimensions 1.2 m × 1.2 m × 1.2 m) and 200 litre steel drums are used as packaging.

Two types of concrete containers are used. The waste from CLAB and the encapsulation plant will be placed in containers with a wall thickness of 10 cm. For the Studsvik waste containers with five 105 litre holes prepared for 85 litre steel drums are used (see Figure 3.1). Some of the steel drums contains inner containers with thick or thin walls.

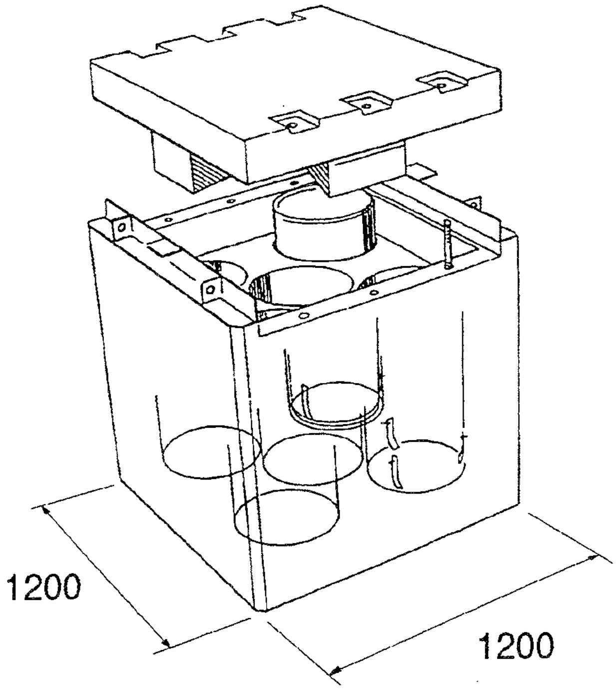

Figure 3.1 *Packaging used for solid intermediate level waste (refuse and scrap), plutonium waste and tritium waste. Five double lid steel drums are placed in five holes in the concrete container.*

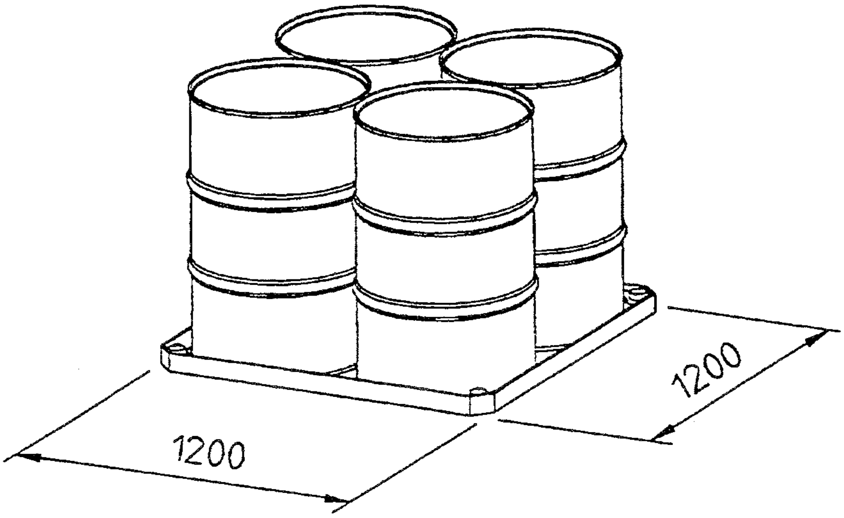

Figure 3.2 *Packaging used for solidified sludge, ashes, refuse and scrap. 200 litre steel drums.*

The 200 litre steel drums (see Figure 3.2) are of two types. Steel drums with mixers are used for solidified sludge and steel drums with inner drums surrounded by concrete for ashes, refuse and scrap.

Amount of waste packages

The total volume required for the waste packages in SFL 3 (sum of the outer volume of the waste packages) has been estimated to be about 5200 m^3. The contribution from Studsvik is about 2400 m^3 and from CLAB and the encapsulation plant together approximately 3800 m^3. The estimated number and volume of packages of different waste types are given in Table 3.1. Some waste types can be treated in alternative ways. In the table the most plausible alternative for each category are given and the alternatives are indicated.

Table 3.1 Waste types in SFL 3, number of packages and total volume of packages (chosen alternatives are underlined).

Waste type	Number of packages	Volume (m^3)
Studsvik		
Concrete containers with ILW	400	691
Concrete containers with plutonium waste	50/$\underline{100}$[a)]	86/$\underline{172}$[a)]
Concrete containers with tritium waste	2	3
Steel drums with solidified sludge	600	128
Steel drums with ashes containing uranium	147	31
Steel drums with refuse and scrap containing plutonium	400	86
Steel drums with refuse and scrap without plutonium	1000	214
Steel containers with cadmium scrap	$\underline{12}$/12[b)]	$\underline{9}$/21[b)]
Stainless steel boxes with plutonium waste	$\underline{0}$[a)]/11	$\underline{0}$[a)]/55
Concrete boxes with plutonium waste	29	58
Steel container with decommissioning waste	$\underline{1}$/1[b)]	$\underline{1}$/2[b)]
Steel drums with decommissioning waste	100	21
CLAB and the encapsulation plant		
Concrete moulds with ion-exchange resins	1800	3110
Concrete moulds with scrap	360	622
Total		**5149**[c)]

a) The plutonium waste in stainless steel boxes may possibly be reconditioned and repacked in concrete containers. This will generate 50 additional concrete containers and increase the volume by 86 m^3.

b) The cadmium scrap and the decommissioning waste in steel containers may possibly be packed in concrete containers.

c) The underlined alternatives have been chosen when summed up.

Radionuclide inventory

The total activity in SFL 3 at the time of repository closure (year 2040) has been estimated to about $3 \cdot 10^{15}$ Bq. Initially the activity is dominated by the H-3 content in one of the Studsvik waste types, tritium immobilised as titaniumhydride [*Holtslander and Miller, 1982*]. Hereafter the activity is dominated by Ni-63 and Ni-59 in the operational waste from CLAB and the encapsulation plant. The activity content of dominant radionuclides in SFL 3 as a function of time is presented in Figure 3.3 and the content of all considered radionuclides is given in Table 3.7.

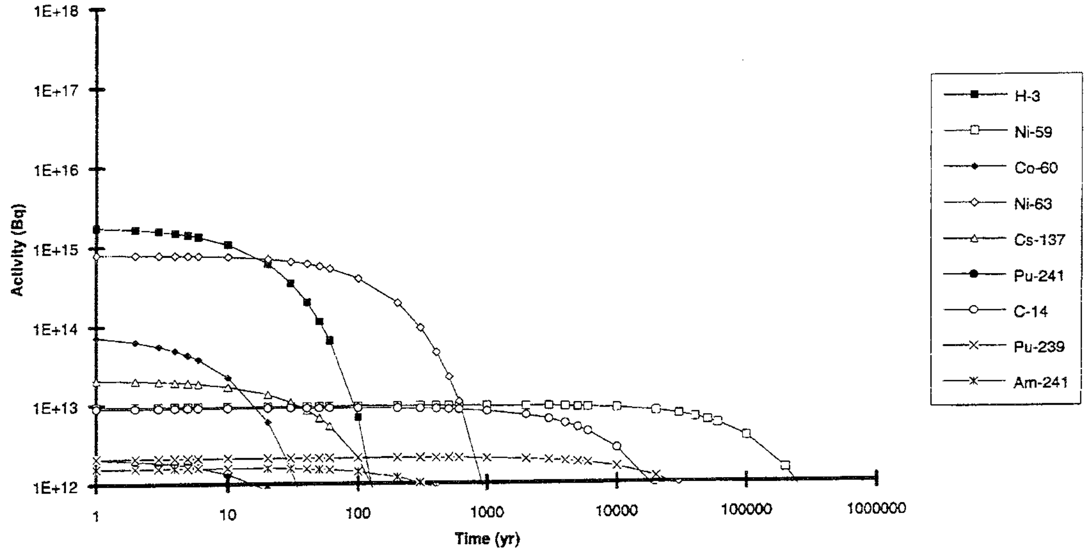

Figure 3.3 Activity content of different radionuclides in SFL 3 as function of time (time zero is at year 2040).

3.2 SFL 4 waste

General

In SFL 4 low-level decommissioning waste from the central interim storage for spent fuel, CLAB and the encapsulation plant, as well as transport casks, transport containers and fuel storage canisters will be disposed. The storage volume required is about 10 000 m^3 of which 2200 m^3 is decommissioning waste, 6600 m^3 is storage canisters from CLAB, and 1200 m^3 is transport casks and containers. The waste will only contain metals and concrete.

Raw waste description and origin

The decommissioning waste from CLAB is divided into two waste categories: steel components and concrete. The steel components are mainly parts from the cooling and clean-up system in the storage pool, i.e. tubing, pumps and tanks. The concrete is from the buildings, since concrete has to be chipped off from rooms potentially classified as contaminated.

The decommissioning waste from the encapsulation plant is foreseen to be of the same type as from CLAB, but the amounts will be smaller.

The transport casks and transport containers are used during transportation of high- and intermediate-level waste between the power plant, the interim storage and the final disposal site. The transport casks and transport containers will, as one option, be disposed of in SFL 4.

The storage canisters holding the fuel assemblies in the storage pools in CLAB are also foreseen to be allocated to SFL 4.

Packaging and conditioning

The waste packaging for the decommissioning waste and fuel storage canisters has not been finally designed yet. In this study a waste packaging denoted "unit vessel" is used. The unit vessel is a cubic carbon steel vessel with a side length of 2.4 m (see Figure 3.4). The vessels will not be backfilled.

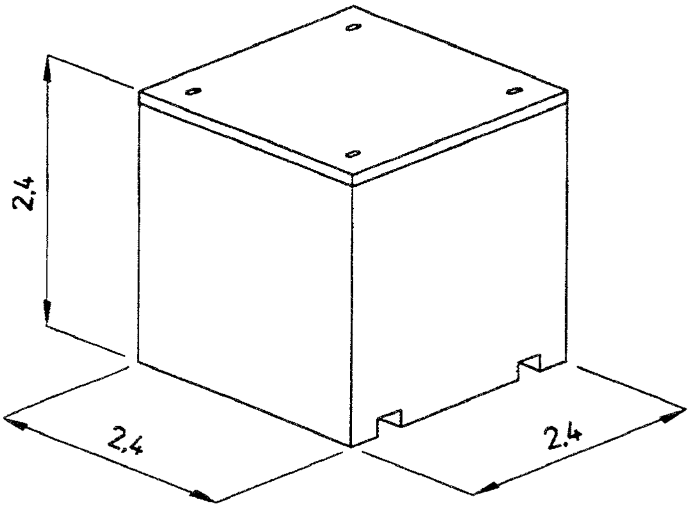

Figure 3.4 Packaging used for decommissioning waste (unit vessel).

Amount of waste packages

The total volume required for the waste in SFL 4 (sum of the outer volume of the waste packages) is about 10 000 m^3. The estimated number of packages and the outer volume required by the different waste types is given in Table 3.2.

Table 3.2 Waste types in SFL 4, number of packages and total volume of packages.

Waste type	Number of packages	Volume (m^3)
Waste in unit vessels		
Components from CLAB	98	1350
Concrete from CLAB	53	730
Fuel storage canisters from CLAB	480	6630
Components from the encapsulation plant	6	80
Concrete from the encapsulation plant	3	40
Total (waste in unit vessels)	*640*	*8830*
Transport casks and transport containers		
Transport casks for spent fuel	10	100
Transport casks for core components	2	20
Transport casks for spent fuel canisters	40	400
Transport containers for ILW	25	680
Total (transport casks and containers)	*77*	*1200*
Total		**10030**

Radionuclide inventory

The total activity in SFL 4 has been estimated to about $2 \cdot 10^{14}$ Bq and is determined by the surface contamination on the spent fuel storage canisters. During the first twenty years the radionuclides Fe-55 and Co-60 dominate and thereafter Ni-63 followed by Ni-59. The activity content as a function of time for the dominating radionuclides are presented in Figure 3.5 and the content of all foreseen radionuclides in the SFL 4 waste is given in Table 3.7.

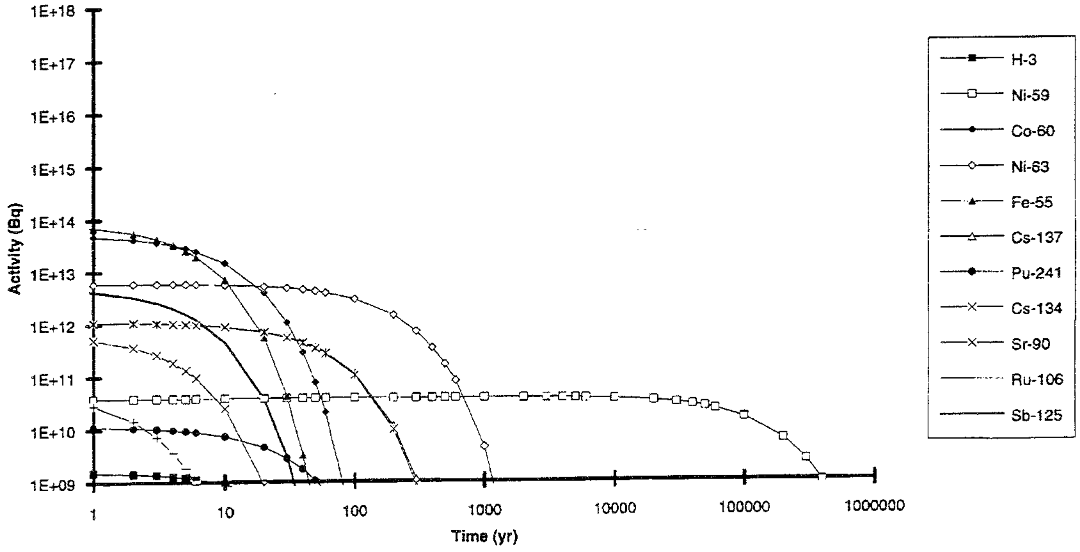

Figure 3.5 Activity content of different radionuclides in SFL 4 as function of time (time zero is at year 2040).

3.3 SFL 5 waste

General

SFL 5 is intended for metallic reactor core components and internal parts with high specific activity; both operational and decommissioning waste. The storage volume required is about 9600 m^3.

Raw waste description and origin

The waste is produced both during operation and at decommissioning of the nuclear power units. The majority of the waste is stainless steel components. Additional materials in the waste are boron steel, boron carbide, hafnium, Zircaloy, inconel and boron glass. The waste in SFL 5 will not contain any organic materials.

The waste originates from the central part of the reactor, close to the core or inside the core, where neutron irradiation of different structural materials create induced activity. The induced activity decreases with the distance from the core and at a few meters distance from the core the crud activity dominates. Before the repository is built used reactor components are kept in interim storage at the power plants and in the central interim storage for spent fuel (CLAB).

Packaging and conditioning

The waste packaging used in SFL 5 is a container made of reinforced concrete with an inner steel cassette. The cassettes with the waste will be placed inside the concrete container and a lid will be put on. The lid is bolted tight and cement will be injected through the lid until the space between the cassette and the concrete container is filled.

The dimensions of the concrete containers are 1.2 × 1.2 × 4.8 m (see Figure 3.6).

Two types of inner steel cassettes is proposed to be used. Most of the waste will be stored in metal scrap cassettes. This cassette has an eight-edged cross section, with a width of 0.761 m and a length of 4.56 m. The other type of cassette is a CLAB cassette which is of the same type as the storage canisters for fuel assemblies used in CLAB.

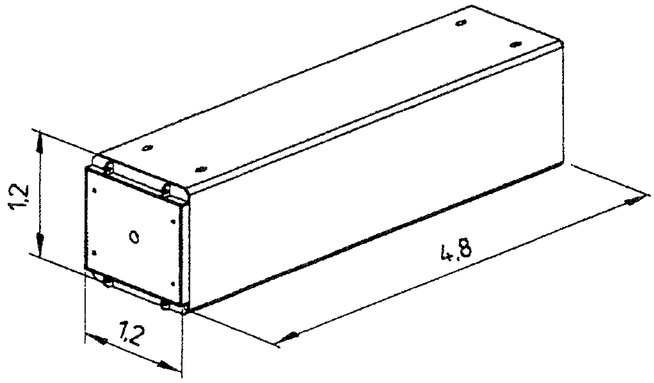

Figure 3.6 Packaging used for reactor internals and components.

Amount of waste packages

In Table 3.3 a prognosis of the number and total volume of packages of different waste types in SFL 5 are given. The estimated total volume of almost 9600 m^3 is the sum of the outer volume of the packages. The number of packages differs substantially between different waste types. The waste type which needs the largest waste volume is the one that comprises the control rods from the BWRs.

Table 3.3 Waste types in SFL 5, number of packages and total volume of packages.

Waste type	Number of packages	Volume (m^3)
BWR		
Core grid	128	885
Moderator tank	81	560
Moderator tank cover	189	1306
Core spray with support	108	746
Control rod guide tubes	45	311
Instrumental tubes	9	62
Boron plates	20	138
Control rods	419	2896
Used parts	4	28
Compacted fuel boxes	14	97
Transition pieces from fuel boxes	1	7
Fuel boxes and SRM/IRM-detectors	11	76
Fuel boxes and PRM-detectors and tubes	12	83
PRM-detectors and tubes	57	394
TIP-detectors	19	131
PWR		
Reactor tank	63	435
Rod bundles	51	353
Internal parts	150	1037
Decommissioning waste from Studsvik	1	7
Total	1382	9552

Radionuclide inventory

The total activity content in the SFL 5 waste has been estimated to about $1 \cdot 10^{17}$ Bq at the year 2040. The activity is determined by the induced activity in the core components and the internal parts. At the time of repository closure the content of Co-60 has decreased due to decay and the activity is dominated by the nickel isotopes. Initially Ni-63 dominates and after 1000 years Ni-59 is the dominant radionuclide, see Figure 3.7. The activity of H-3 in the BWR control rods is of the same magnitude as the Co-60 activity at the time of repository closure. The estimated activity content of all considered radionuclides in the waste is presented in Table 3.7.

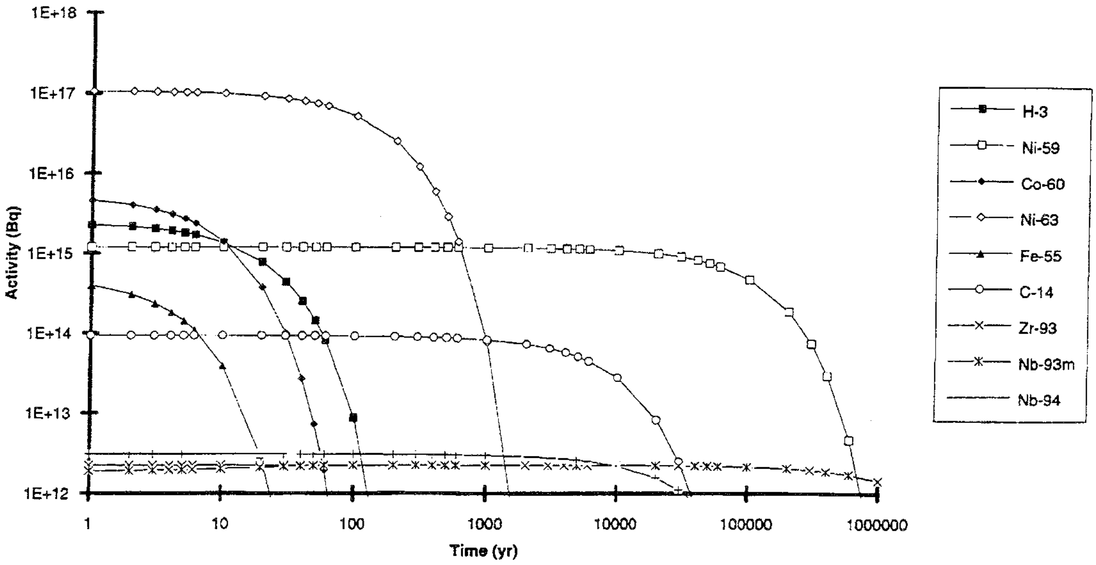

Figure 3.7 Activity content of different radionuclides in SFL 5 as function of time (time zero is at year 2040).

3.4 Total inventory of waste

In this section a summary of the amounts of materials and activities in the different repository parts is given. In Table 3.4 a summary of the total waste volumes (sum of the outer volume of the packages) in the different repository parts is given. The total volume is approximately 25 000 m^3.

The amounts (tonnes) of different materials in the waste are given in Table 3.5. The dominating waste materials are steel, 12 000 tonnes, and concrete/cement, 3800 tonnes. Waste containing organic material is allocated to SFL 3 and the prognosis is that there will be a total of about 400 tonnes of organic material in SFL 3. In addition to the waste materials there is steel and concrete in the packaging, see Table 3.6.

Table 3.4 Volumes in SFL 3-5 (sum of the outer volume of the packages).

Repository part	Volume (m^3)
SFL 3	5200
SFL 4	10000
SFL 5	9600
Total	24800

Table 3.5 Amounts (tonnes) of different materials in SFL 3-5 waste (excluding packagings).

Material	SFL 3	SFL 4	SFL 5	Total
Metals				
Steel	217	8800[b)]	1900	10917
Aluminium	51			51
Lead	7.5			7.5
Cadmium	1.2			1.2
Brass and copper	7.5			7.5
Beryllium	0.3		0.3	0.6
Hafnium			4	4
Zircaloy			30	30
Organics				
Ion-exchange resins	270			270
Cellulose	46			46
Plastic/Rubber	67			67
Others	2.8			2.8
Concrete/Cement[a)]	2360	850		3210
Others				
Ferrocyanide precipitates	4.6			4.6
Uranium contaminated ashes	2.9			2.9
Total	3038	9650	1934	14622

a) solidified waste and contaminated concrete.
b) including transport casks and transport containers

Table 3.6 Amounts (tonnes) of materials in SFL 3-5 packagings.

Material	SFL 3	SFL 4	SFL 5	Total
Lead[a)]			124	124
Steel[b)]	760	1000	1500	3260
Concrete [c)]	7500		18900	26400

a) shielding for TIP-detectors
b) including reinforcement in concrete containers
c) including backfill in packagings

Radionuclide inventory

The estimated radionuclide inventory at the time of repository closure (year 2040) is given in Table 3.7 for the different repository parts and for the whole repository. The total activity content in SFL 3-5 is about $1 \cdot 10^{17}$ Bq.

The initial activity content is dominated by Ni-63 followed by the radionuclides Co-60, H-3, Ni-59 and C-14. More than 90 % of the total initial activity originates from the waste in SFL 5 and is determined by Ni-63. In Figure 3.8 the total activity content in the different repository parts as a function of time is presented. The total activity content in the repository is determined by the activity in SFL 5 and by the nickel isotopes. However, other radionuclides may be more or equally important from safety point e.g cesium-isotopes and actinides. The content of these radionuclides in the repository is dominated by the content in the SFL 3 waste.

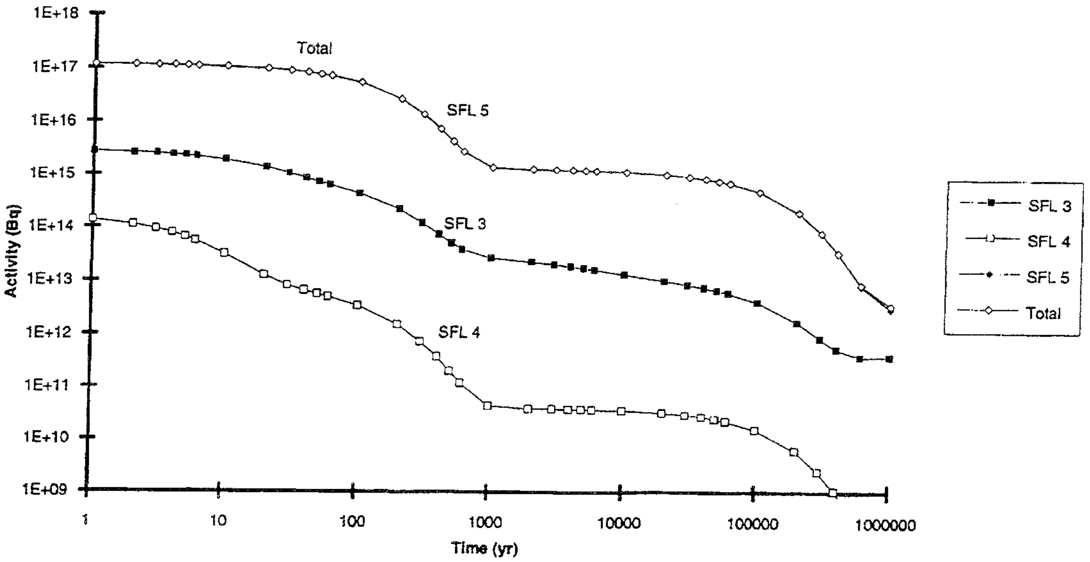

Figure 3.8 Activity content of the different repository parts as function of time (time zero is at year 2040).

To get a measure of the radiotoxicity of the waste to be disposed of in SFL 3-5 the dose conversion factors for ingestion from [*NRPB-R245, 1991*] have been used to convert the activity content of different radionuclides given in Bq to potential doses in Sv. These potential doses can be used to compare radiological hazards from different wastes, repository parts and disposal systems.

Radiotoxicity [Sv] = Activity content [Bq] × Dose conversion factor [Sv/Bq]

The estimated radiotoxicities for individual radionuclides in the different repository parts at repository closure are presented in Table 3.8. The total radiotoxicity in SFL 3-5 is initially determined by the content of Co-60 and Ni-63 in the SFL 5 waste. The initial radiotoxicities of the SFL 3 and SFL 4 waste is more than one order of magnitude lower than the SFL 5 waste. At longer times the radiotoxicity is dominated by the long-lived actinides Am-241, Pu-239 and Pu-240 in the SFL 3 waste. In Figure 3.9 the contribution from the different repository parts to the total radiotoxicity as a function of time is presented. The total radiotoxicity of the SFL 4 waste decreases with more than four order of magnitudes under the first thousand years. The radiotoxicity of the SFL 3 waste is more or less constant under the same period and exceeds the radiotoxicity of SFL 5 waste approximately at thousand years.

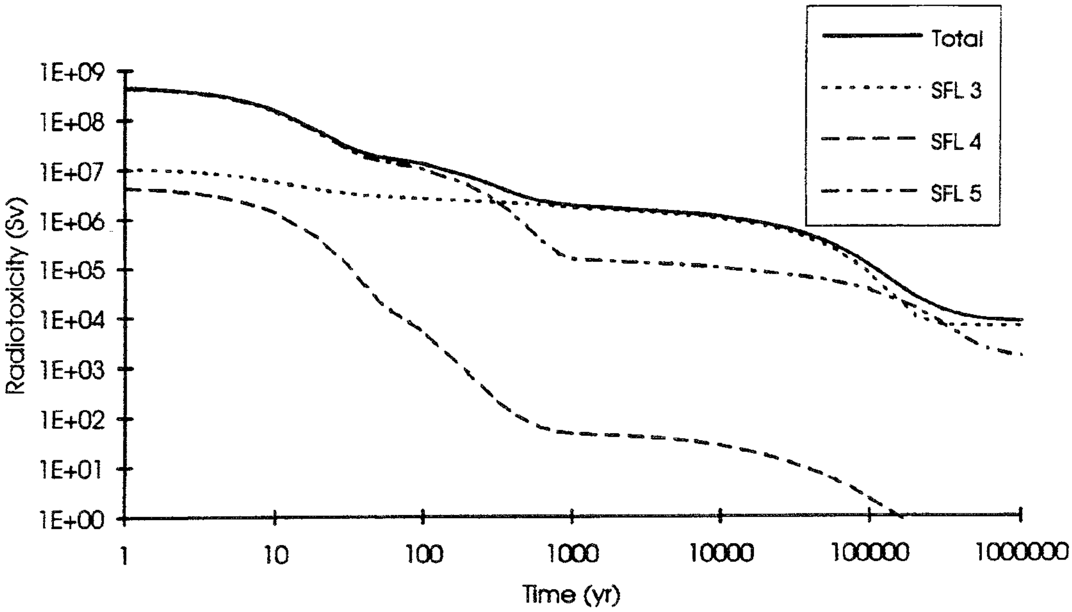

Figure 3.9 Radiotoxicity of the different repository parts as function of time (time zero is at year 2040).

Toxic waste inventory

In the SFL 3 waste some of the metals present in waste from Studsvik are toxic such as lead, cadmium and beryllium. In fact some of the radionuclide elements are also chemotoxic such as uranium and plutonium. In the SFL 5 waste, lead is used as shielding for the TIP-detectors and beryllium is found in the metallic waste from decommissioning of the research reactor (R2) at Studsvik. The SFL 4 waste comprises mainly of steel components.

3.5 Additional comments

The waste to SFL 3-5 has many similarities with the waste to SFR. Strictly taken, not all of the waste forms destined for SFL 3, 4 and 5 falls into the category of long-lived waste. In fact, only the waste that comes from Studsvik, the core components and the reactor internals are to be classified as long-lived. Operational waste and later decommissioning waste from CLAB and the encapsulation plant could in principle be disposed of in SFR. However, SFL 3-5 is intended to receive all LLW and ILW that arises in the post-closure period of SFR.

It should be kept in mind that part of the waste foreseen for SFL 3-5 in this summary may in fact turn out to be possible to decontaminate and thereby exempt this waste from regulatory control.

At Studsvik fissile and fertile U, Pu and Th are stored. It is possible that some of this material has to be disposed of in SFL if it cannot be reused. However, until this has been decided, no such material is included in the waste inventory.

The uncertainties in the estimates of waste amounts and radionuclide inventory are large especially regarding old conditioned waste from Studsvik. Ongoing projects at Studsvik will however give more accurate information about the waste.

Table 3.7 Radionuclide inventory in SFL 3-5 at year 2040 given in Bq.

Nuclide*)	**Half life**	**SFL 3**	**SFL 4**	**SFL 5**	**Total**
H-3	$1.2\cdot10^{1}$	$1.9\cdot10^{15}$	$1.6\cdot10^{9}$	$2.4\cdot10^{15}$	$4.3\cdot10^{15}$
Be-10	$1.6\cdot10^{6}$			$1.4\cdot10^{11}$	$1.4\cdot10^{11}$
C-14	$5.7\cdot10^{3}$	$9.0\cdot10^{12}$		$9.3\cdot10^{13}$	$1.0\cdot10^{14}$
Fe-55	$2.7\cdot10^{0}$	$6.5\cdot10^{11}$	$9.3\cdot10^{13}$	$5.1\cdot10^{14}$	$6.1\cdot10^{14}$
Co-60	$5.3\cdot10^{0}$	$8.3\cdot10^{13}$	$5.4\cdot10^{13}$	$5.2\cdot10^{15}$	$5.3\cdot10^{15}$
Ni-59	$7.5\cdot10^{4}$	$9.4\cdot10^{12}$	$3.8\cdot10^{10}$	$1.2\cdot10^{15}$	$1.2\cdot10^{15}$
Ni-63	$9.6\cdot10^{1}$	$8.0\cdot10^{14}$	$6.0\cdot10^{12}$	$1.1\cdot10^{17}$	$1.1\cdot10^{17}$
Sr-90 a	$2.9\cdot10^{1}$	$9.9\cdot10^{11}$	$1.1\cdot10^{12}$	$3.8\cdot10^{11}$	$2.5\cdot10^{12}$
Zr-93	$1.5\cdot10^{6}$	$4.7\cdot10^{7}$	$5.4\cdot10^{7}$	$2.2\cdot10^{12}$	$2.2\cdot10^{12}$
Nb-93m	$1.4\cdot10^{1}$	*$1.9\cdot10^{7}$*	$2.6\cdot10^{8}$	$1.9\cdot10^{12}$	$1.9\cdot10^{12}$
Nb-94	$2.0\cdot10^{4}$	$9.2\cdot10^{9}$	$5.5\cdot10^{8}$	$3.1\cdot10^{12}$	$3.1\cdot10^{12}$
Mo-93	$3.5\cdot10^{3}$	$1.8\cdot10^{7}$	$5.4\cdot10^{7}$	$3.8\cdot10^{7}$	$1.1\cdot10^{8}$
Tc-99	$2.1\cdot10^{5}$	$3.0\cdot10^{9}$	$5.4\cdot10^{7}$	$3.3\cdot10^{11}$	$3.4\cdot10^{11}$
Ru-106 b	$1.0\cdot10^{0}$	$6.5\cdot10^{5}$	$5.7\cdot10^{10}$	$7.0\cdot10^{1}$	$5.7\cdot10^{10}$
Sb-125 c	$2.8\cdot10^{0}$	$9.2\cdot10^{10}$	$5.4\cdot10^{12}$	$2.1\cdot10^{9}$	$5.5\cdot10^{12}$
I-129	$1.6\cdot10^{7}$	$1.9\cdot10^{7}$	$4.4\cdot10^{5}$	$3.1\cdot10^{5}$	$1.9\cdot10^{7}$
Cs-134	$2.1\cdot10^{0}$	$3.6\cdot10^{9}$	$7.1\cdot10^{11}$	$2.1\cdot10^{7}$	$7.2\cdot10^{11}$
Cs-135	$2.3\cdot10^{6}$	$1.9\cdot10^{8}$	$1.6\cdot10^{7}$	$1.1\cdot10^{7}$	$2.2\cdot10^{8}$
Cs-137 d	$3.0\cdot10^{1}$	$2.1\cdot10^{13}$	$1.1\cdot10^{12}$	$3.9\cdot10^{11}$	$2.3\cdot10^{13}$
Eu-152	$1.3\cdot10^{1}$	$2.9\cdot10^{10}$			$2.9\cdot10^{10}$
Eu-154	$8.8\cdot10^{0}$	$5.6\cdot10^{9}$			$5.6\cdot10^{9}$
Eu-155	$5.0\cdot10^{0}$	$4.0\cdot10^{9}$			$4.0\cdot10^{9}$
Pb-210 h	*$2.2\cdot10^{1}$*	*$7.8\cdot10^{11}$*			*$7.8\cdot10^{11}$*
Ra-226 e	$1.6\cdot10^{3}$	$9.8\cdot10^{11}$			$9.8\cdot10^{11}$
Ac-227 g	*$2.2\cdot10^{1}$*	*$4.4\cdot10^{6}$*			*$4.4\cdot10^{6}$*
Th-230	*$7.7\cdot10^{4}$*	*$2.7\cdot10^{3}$*		*$1.3\cdot10^{0}$*	*$2.7\cdot10^{3}$*
Th-232 f	$1.4\cdot10^{10}$	$3.0\cdot10^{9}$			$3.0\cdot10^{9}$
Pa-231	*$3.3\cdot10^{4}$*	*$8.7\cdot10^{6}$*			*$8.7\cdot10^{6}$*
U-234	*$2.4\cdot10^{5}$*	*$1.5\cdot10^{7}$*		*$9.4\cdot10^{3}$*	*$1.5\cdot10^{7}$*
U-235	$7.0\cdot10^{8}$	$8.2\cdot10^{9}$			$8.2\cdot10^{9}$
U-236	*$2.3\cdot10^{7}$*	*$6.6\cdot10^{5}$*		*$2.4\cdot10^{1}$*	*$6.6\cdot10^{5}$*
U-238	$4.5\cdot10^{9}$	$2.0\cdot10^{10}$			$2.0\cdot10^{10}$
Np-237	*$2.1\cdot10^{6}$*	*$2.5\cdot10^{7}$*		*$1.3\cdot10^{3}$*	*$2.5\cdot10^{7}$*
Pu-238	$8.8\cdot10^{1}$	$1.3\cdot10^{11}$	$1.8\cdot10^{8}$	$9.8\cdot10^{7}$	$1.3\cdot10^{11}$
Pu-239	$2.4\cdot10^{4}$	$2.1\cdot10^{12}$	$3.4\cdot10^{7}$	$2.4\cdot10^{7}$	$2.1\cdot10^{12}$
Pu-240	$6.5\cdot10^{3}$	$4.6\cdot10^{11}$	$3.9\cdot10^{7}$	$2.7\cdot10^{7}$	$4.6\cdot10^{11}$
Pu-241	$1.4\cdot10^{1}$	$2.2\cdot10^{12}$	$1.2\cdot10^{10}$	$1.9\cdot10^{9}$	$2.2\cdot10^{12}$
Am-241	$4.3\cdot10^{2}$	$1.6\cdot10^{12}$	$1.2\cdot10^{7}$	$2.1\cdot10^{8}$	$1.6\cdot10^{12}$
Am-243	$7.4\cdot10^{3}$	$9.0\cdot10^{8}$	$3.9\cdot10^{6}$	$2.7\cdot10^{6}$	$9.1\cdot10^{8}$
Cm-243	$2.9\cdot10^{1}$	$4.2\cdot10^{8}$	$7.4\cdot10^{7}$	$2.6\cdot10^{7}$	$5.2\cdot10^{8}$
Cm-244	$1.8\cdot10^{1}$	$9.6\cdot10^{9}$	$2.2\cdot10^{7}$	$4.8\cdot10^{6}$	$9.6\cdot10^{9}$
Total		$2.8\cdot10^{15}$	$1.6\cdot10^{14}$	$1.2\cdot10^{17}$	$1.2\cdot10^{17}$

*) The estimated inventory of nuclides formed by ingrowth up to the time of repository closure is given in italic.

a Sr-90 is in equilibrium with the daughter nuclide Y-90

b Ru-106 is in equilibrium with the daughter nuclide Rh-106

c Sb-125 is in equilibrium with the daughter nuclide Te-125m

d Cs-137 is in equilibrium with the daughter nuclide Ba-137m

e Ra-226 is in equilibrium with daughter nuclides until Pb-210

f Th-232 is in equilibrium with daughter nuclides

g Ac-227 is in equilibrium with daughter nuclides

h Pb-210 is in equilibrium with daughter nuclides

Table 3.8 Compilation of dose conversion factors for ingestion from [NRPB-R245, 1991] and radiotoxicities in SFL 3-5 at year 2040 given in Sv.

Nuclide*)	Dose factor (Sv/Bq)	SFL 3	SFL 4	SFL 5	Total
H-3	$1.80\cdot10^{-11}$	$3.4\cdot10^{4}$	$2.9\cdot10^{-2}$	$4.3\cdot10^{4}$	$7.7\cdot10^{4}$
Be-10	$1.80\cdot10^{-9}$			$2.6\cdot10^{2}$	$2.6\cdot10^{2}$
C-14	$5.60\cdot10^{-10}$	$5.0\cdot10^{3}$		$5.2\cdot10^{4}$	$5.7\cdot10^{4}$
Fe-55	$4.00\cdot10^{-10}$	$2.6\cdot10^{2}$	$3.7\cdot10^{4}$	$2.1\cdot10^{5}$	$2.4\cdot10^{5}$
Co-60	$9.20\cdot10^{-8}$	$7.6\cdot10^{6}$	$5.0\cdot10^{6}$	$4.8\cdot10^{8}$	$4.9\cdot10^{8}$
Ni-59	$6.80\cdot10^{-11}$	$6.4\cdot10^{2}$	$2.6\cdot10^{0}$	$8.1\cdot10^{4}$	$8.2\cdot10^{4}$
Ni-63	$1.90\cdot10^{-10}$	$1.5\cdot10^{5}$	$1.1\cdot10^{3}$	$2.0\cdot10^{7}$	$2.0\cdot10^{7}$
Sr-90	$2.80\cdot10^{-8}$	$2.8\cdot10^{4}$	$3.1\cdot10^{4}$	$1.1\cdot10^{4}$	$6.9\cdot10^{4}$
Zr-93	$1.00\cdot10^{-9}$	$4.7\cdot10^{-2}$	$5.4\cdot10^{-2}$	$2.2\cdot10^{3}$	$2.2\cdot10^{3}$
Nb-93m	$2.10\cdot10^{-10}$	*$4.0\cdot10^{-3}$*	$5.4\cdot10^{-2}$	$3.9\cdot10^{2}$	$3.9\cdot10^{2}$
Nb-94	$2.30\cdot10^{-9}$	$2.1\cdot10^{1}$	$1.3\cdot10^{0}$	$7.1\cdot10^{3}$	$7.1\cdot10^{3}$
Mo-93	$2.60\cdot10^{-10}$	$4.7\cdot10^{-3}$	$1.4\cdot10^{-2}$	$9.8\cdot10^{-3}$	$2.9\cdot10^{-2}$
Tc-99	$6.70\cdot10^{-10}$	$2.0\cdot10^{0}$	$3.6\cdot10^{-2}$	$2.2\cdot10^{2}$	$2.3\cdot10^{2}$
Ru-106	$1.10\cdot10^{-8}$	$7.1\cdot10^{-3}$	$6.2\cdot10^{2}$	$7.7\cdot10^{-7}$	$6.2\cdot10^{2}$
Sb-125	$9.80\cdot10^{-10}$	$9.0\cdot10^{1}$	$5.3\cdot10^{3}$	$2.0\cdot10^{0}$	$5.4\cdot10^{3}$
I-129	$1.10\cdot10^{-7}$	$2.0\cdot10^{0}$	$4.9\cdot10^{-2}$	$3.4\cdot10^{-2}$	$2.1\cdot10^{0}$
Cs-134	$1.90\cdot10^{-8}$	$6.8\cdot10^{1}$	$1.4\cdot10^{4}$	$4.0\cdot10^{-1}$	$1.4\cdot10^{4}$
Cs-135	$1.90\cdot10^{-9}$	$3.6\cdot10^{-1}$	$3.1\cdot10^{-2}$	$2.2\cdot10^{-2}$	$4.1\cdot10^{-1}$
Cs-137	$1.30\cdot10^{-8}$	$2.8\cdot10^{5}$	$1.4\cdot10^{4}$	$5.0\cdot10^{3}$	$3.0\cdot10^{5}$
Eu-152	$2.00\cdot10^{-9}$	$5.9\cdot10^{1}$			$5.9\cdot10^{1}$
Eu-154	$3.10\cdot10^{-9}$	$1.7\cdot10^{1}$			$1.7\cdot10^{1}$
Eu-155	$5.30\cdot10^{-10}$	$2.1\cdot10^{0}$			$2.1\cdot10^{0}$
Pb-210	*$1.30\cdot10^{-6}$*	*$1.0\cdot10^{6}$*			*$1.0\cdot10^{6}$*
Ra-226	$2.20\cdot10^{-7}$	$2.2\cdot10^{5}$			$2.2\cdot10^{5}$
Ac-227	*$2.20\cdot10^{-6}$*	*$9.6\cdot10^{0}$*			*$9.6\cdot10^{0}$*
Th-230	*$3.50\cdot10^{-7}$*	*$9.4\cdot10^{-4}$*		*$4.6\cdot10^{-7}$*	*$9.4\cdot10^{-4}$*
Th-232	$1.80\cdot10^{-6}$	$5.4\cdot10^{3}$			$5.4\cdot10^{3}$
Pa-231	*$1.40\cdot10^{-6}$*	*$1.2\cdot10^{1}$*			*$1.2\cdot10^{1}$*
U-234	*$3.90\cdot10^{-8}$*	*$5.7\cdot10^{-1}$*		*$3.7\cdot10^{-4}$*	*$5.7\cdot10^{-1}$*
U-235	$3.80\cdot10^{-8}$	$3.1\cdot10^{2}$			$3.1\cdot10^{2}$
U-236	*$3.70\cdot10^{-8}$*	*$2.4\cdot10^{-2}$*		*$8.8\cdot10^{-7}$*	*$2.4\cdot10^{-2}$*
U-238	$3.60\cdot10^{-8}$	$7.3\cdot10^{2}$			$7.3\cdot10^{2}$
Np-237	*$6.40\cdot10^{-7}$*	*$1.6\cdot10^{1}$*		*$8.4\cdot10^{-4}$*	*$1.6\cdot10^{1}$*
Pu-238	$5.10\cdot10^{-7}$	$6.8\cdot10^{4}$	$9.1\cdot10^{1}$	$5.0\cdot10^{1}$	$6.8\cdot10^{4}$
Pu-239	$5.60\cdot10^{-7}$	$1.2\cdot10^{6}$	$1.9\cdot10^{1}$	$1.3\cdot10^{1}$	$1.2\cdot10^{6}$
Pu-240	$5.60\cdot10^{-7}$	$2.6\cdot10^{5}$	$2.2\cdot10^{1}$	$1.5\cdot10^{1}$	$2.6\cdot10^{5}$
Pu-241	$1.10\cdot10^{-8}$	$2.4\cdot10^{4}$	$1.3\cdot10^{2}$	$2.1\cdot10^{1}$	$2.4\cdot10^{4}$
Am-241	$5.70\cdot10^{-7}$	$9.0\cdot10^{5}$	$7.0\cdot10^{0}$	$1.2\cdot10^{2}$	$9.0\cdot10^{5}$
Am-243	$5.70\cdot10^{-7}$	$5.1\cdot10^{2}$	$2.2\cdot10^{0}$	$1.5\cdot10^{0}$	$5.2\cdot10^{2}$
Cm-243	$4.00\cdot10^{-7}$	$1.7\cdot10^{2}$	$3.0\cdot10^{1}$	$1.0\cdot10^{1}$	$2.1\cdot10^{2}$
Cm-244	$3.20\cdot10^{-7}$	$3.1\cdot10^{3}$	$6.9\cdot10^{0}$	$1.5\cdot10^{0}$	$3.1\cdot10^{3}$
Total		$1.2\cdot10^{7}$	$5.1\cdot10^{6}$	$5.0\cdot10^{8}$	$5.2\cdot10^{8}$

*) The estimated inventory of nuclides formed by ingrowth up to the time of repository closure is given in italic.

4 Barrier properties

The SFL 3-5 near-field barrier system is defined as the engineered barriers and the nearest surrounding rock. There is no clearly defined limit between near-field and far-field rock. In this study we define the near-field rock as the part of the host rock that will be influenced by the excavation works. Important engineered barriers are for example waste packaging, structures made of concrete, and backfill in different locations made of porous concrete, bentonite, sand/bentonite mixtures and gravel. The physical and chemical properties of the barrier materials and the chemical influence of the near-field water chemistry are discussed in the following sections. Also a brief discussion of the long-time stability of concrete and bentonite is included. The effects of interaction between concrete and rock or between concrete and bentonite were not included in this prestudy.

4.1 Concrete

The most common cement type in Sweden is Portland cement. The most frequently used is Degerhamn Standard Portland which is a sulphate resistant cement due to its relatively low content of aluminium [*Lagerblad and Trädgård, 1995*]. There exists other types of cement such as blast-furnace slag cements etc. Almost all the cement types are based on Portland cement with different added solids. The same hydration products will be found in all kinds of Portland cements, although the proportions may vary.

The cement clinker of Ordinary Portland Cement (OPC) consists mainly of the following solid phases; tricalcium silicate, dicalcium silicate, tricalcium aluminate and tetracalcium aluminate ferrite. The typical ranges of concentrations in OPC [*Andersson and Allard, 1986*] are presented in Table 4.1. The Degerhamn Standard Portland Cement (SPC) used as construction cement in SFR has also been selected as a reference material in this study. The clinker composition of SPC [*Lagerblad and Trädgård, 1995*] is presented in Table 4.1.

Table 4.1 Clinker composition of Ordinary Portland Cement (OPC) and Degerhamn Standard Portland Cement (SPC).

Clinker component	**OPC[a] weight %**	**SPC[b] weight %**
C_3S	45 - 65	51
C_2S	10 - 30	25
C_3A	5 - 15	1.2
C_4AF	5 - 12	14

Abbreviations: C=CaO, S=SiO_2, A=Al_2O_3, F=Fe_2O_3

[a] [*Andersson et al., 1986*]

[b] [*Lagerblad et al., 1994*]

When the ground cement clinker is mixed with water it hydrates and forms new solid phases. The main hydration reactions for the most important solid clinker phases in OPC are [*Czernin, 1969*]:

C_3S: $2\ C_3S + 6\ H_2O = C_3S_2{\cdot}3H_2O + 3\ Ca(OH)_2$

C_2S: $2\ C_2S + 4\ H_2O = C_3S_2{\cdot}3H_2O + Ca(OH)_2$

C_3A: $C_3A + 6\ H_20 = C_3A{\cdot}6H_2O$

C_4AF: $4\ C_4AF + 2\ Ca(OH)_2 + 10\ H_2O = C_3A{\cdot}6H_2O + C_3F{\cdot}6H_2O$

The hydrated cement forms a gel which also contains some crystalline components. In the solid there is an initially water filled pore system (10 - 15 % of the volume). The gel crystallizes slowly, and gel phases are found after very long time [*Lea, 1970*].

The hydrated cement contains considerable amounts of calcium hydroxide. Calcium silicate, calcium aluminate and calcium ferrite phases are also present.

Various additives are used when cement is mixed with water in order to improve the properties. An addition of a workability aid, e.g. sulfonated melamine-formaldehyde polymer, will presumably be made to all the cements used in a repository. The total amount of additives is usually much less than 3 % of the cement weight [*Andersson and Allard, 1986*].

Concrete is a mixture of cement and sand.The sand is referred to as the aggregate phase. Various silicate minerals and quartz are generally represented in the aggregate phase and various grain sizes are used from fine sand to crushed rock.

Experimental determinations of pore water composition have been made, mainly for Portland cement [*Lagerblad and Trädgård, 1995*]. One of these studies compares the compositions of a number of different cements [*Andersson 1983, Andersson et al., 1989*]. Analysis of pore water components in leached cement samples from a 70 year old hydropower dam have also been reported [*Gjørv and Havdal, 1982*]. Pore water data for OPC and the leached cement are given in Table 4.2.

The highly soluble alkali hydroxides in fresh concrete will cause a high pH (about 13.5) in the pore water. If the alkali hydroxides are leached out and transported away the pH will be controlled by the dissolution of portlandite, $Ca(OH)_2$, giving a pH of about 12.5. The porosity will be about 0.15.

If the cement phase is depleted of portlandite by leaching, a pH in the range of 12.5 - 10.5 is expected due to the presence of the CSH-phases in the hydrated cement. The reactions controlling the pH are the incongruent dissolution of the CSH-gel and the formation of secondary phases. A successive decrease in pH is expected due to changes in the C/S ratio from 1.7 (giving a pH of 12.5) to 0.85 (giving a pH of 11) [*Atkins and Glasser, 1992*]. This period is then followed by congruent dissolution of the remaining CSH-gel, buffering the pH at a value of about 10. The porosity will be around 0.3 [*Höglund and Bengtsson, 1991, Andersson and Allard, 1986*].

Table 4.2 Composition of cement porewaters. Ion concentrations in mg/l.

	Fresh cement		Leached cement	
	OPC[a]	OPC[b]	Sample1[c]	Sample2[c]
Age	3 months	10 months	70 years	70 years
w/c[d]	0.4	0.5	-	-
Na	644	1500	1500	260
K	3245	6300	1530	180
Ca	36	90	92	570
Mg	-	0.2	-	-
Al	1.1	<5	-	-
Si	22	<6	-	-
Fe	-	0.5	-	-
I, calc[e]	-	0.23		
Eh[f]		139		
pH	13.1	13.4	12.9	12.6

a *[Lagerblad and Trädgård, 1995]*
b *[Andersson, 1983]*
c Two different samples of 70 years old cement *[Gjørv and Havdal, 1982]*.
d w/c = water/cement ratio
e ionic strength, calculated according to $I = 1/2\Sigma c_i z_i^2$
f potential measured with platinum electrode, mV

4.2 Bentonite and sand/bentonite mixtures

Bentonite is a swelling clay due to its content of the smectite clay mineral montmorillonite. Montmorillonite has a considerable cation exchange capacity with generally sodium and/or calcium in the exchange positions. Wyoming bentonite has a high content of sodium montmorillonite which gives the clay good swelling properties. The Wyoming bentonite MX-80, consists to a large extent (85%) of a size fraction that is smaller than 2 μm. Bentonite contain other minerals too, such as quartz, feldspars, other clay minerals, accessory minerals such as carbonates, sulphides etc. and trace amounts of for example organic materials. The average mineral composition of the Wyoming bentonite MX-80 is given in Table 4.3.

Table 4.3 Mineralogical Composition of MX-80 Bentonite [Müller-Vonmoos and Kahr, 1983].

Mineral	Content (%)
Montmorillonite	75
Quartz	15.2
Feldspar	5-8
Carbonate	1.4
Mica	<1
Kaolinite	<1
Pyrite	0.3
Organic Carbon	0.4
Other Minerals	2

Montmorillonite is a layered silicate mineral that may be derived from a pyrophyllite structure by the insertion, between the triple layers of the pyrophyllite, of sheets of molecular water containing free cations, coupled with substitution of Al for Si in the framework sheets [*Hurlbut, 1971*]. The free cations in the water layer can be Na, K, Ca, and Mg. The ions in the interlayer positions are exchangeable and may influence the pore water composition. Weathering of the montmorillonite would give an addition of more strongly bound metal ions, e.g. silica, and aluminum, to the solution.

The bentonite is likely to buffer a water solution, at 25 C, to a pH-value within the range of 7 to 10, and to an Eh value in the range from -0.3 to -0.2 V [*Wanner et al., 1994, Johnston et al., 1984*]. Experimental values in these ranges have also been obtained [*Rantanen et al., 1987*]. The low Eh values may be attributed to the content of sulphide, ferrous minerals and organic material, cf. Table 4.3.

Not only pure bentonite but also a mixture of sand and bentonite is considered as backfill. However, no major chemical effect such as different pH, Eh or ionic strength is expected due to the presence of sand in the bentonite since the clay acts as a redox and pH buffer.

The equilibrium concentrations of carbonate, calcium and sodium, as well as the pH-value of water in contact with bentonite can be expected to be governed by several chemical reactions. It is of significance that bentonite contains carbonate minerals and some organic material. Dissolution of the carbonate minerals as well as microbial degradation of organic components may occur and cause an increase in dissolved carbonate. The calcium concentrations will be related to the solubility of calcite ($CaCO_3$).

The smectite clay mineral montmorillonite has a high ion exchange capacity and therefore the ion exchange reactions coupled with calcite dissolution will strongly influence the dissolved carbonate, sodium, calcium and pH according to the following reaction:

$$2\,Na(ex) + CaCO_3(s) + H_2O \rightarrow Ca(ex) + 2\,Na^+ + HCO_3^- + OH^-$$

Other cations, like hydrogen, potassium and magnesium ions, will also participate in ion exchange reactions in the sodium bentonite. Measurements have shown that the diffusivity of Na and Ca ions is relatively high in the clay (D_a = 3-5$\cdot 10^{-11}$ m^2/s [*Albinsson et al., 1993*]), which indicates that the sorption of Na^+ and Ca^{2+} in compacted bentonite is very low. The low uptake of Ca^{2+} indicates that the transformation of the sodium bentonite in contact with concrete to its calcium form is minor during the first leaching when pH is about 13.5.

The properties of bentonite may be influenced by the high pH of concrete leachates. Possible reactions are crystal lattice transformations or formation of zeolites, silica and various CHS-phases by smectite lattice breakdown. The rates and yields of these reactions are not well known. In fact, it has been argued that reactions between hydroxyl ions and bentonite would require elevated temperature [*Andersson and Allard, 1986*]. If this is true, there will be no hydrolysis of bentonite due to concrete leachates in the SFL 3-5 repository because the temperature will always be relatively low. However, the conservative approach to assume that reactions take place may also be warranted until more results are available from this field of studies.

The initially Na-saturated bentonite could be converted to illite by replacement of sodium by potassium. Illite is a brittle mineral with little or no swelling property and by decreasing the clay's plasticity, the engineered barrier could, due to cracking, lose some of its hydraulic resistance. This conversion process is greatly enhanced by elevated temperature. At a temperature of 50°C and a potassium concentration of 3 M, 50% of the smectite (bentonite) is converted to illite in 10^5-10^6 years. At 12°C, this process would take more than 10^6 years [*Pusch, 1994*].

A likely ion exchange process in bentonite is the replacement of Na by Ca. This process causes some loss of swelling pressure [*Andersson and Allard, 1986*]. Exchange of Na for Ca is expected to gain importance when the leaching of the alkali hydroxides in the concrete is completed and the concentration in cement porewater of Ca^{2+} starts to increase and Na^+ decreases.

4.3 The chemical influence of near-field materials

The water chemistry in the engineered barriers is initially characterized by the water and humidity already present, the intruding groundwater, air trapped in the voids after sealing and material present in each barrier.

Undisturbed groundwater is expected to have a pH of 6-9 and an Eh of -0.30 to -0.15 V. Geochemical reactions such as ion exchange and weathering of minerals normally control the composition and pH of the groundwater. Iron and sulphide minerals reduce the redox potential. These reactions can also involve organic material, (c.f. 5.3).

The pore waters in concrete structures, porous concrete backfill and concrete containers can initially have a pH as high as 13.5. Due to leaching of the concrete the pH will decrease with time.

The pore water in sand/bentonite barriers is strongly influenced by reactions in the bentonite and for example pH will be effectively controlled by reactions with the bentonite minerals. Bentonite has a buffering capacity for pH and Eh, (c.f. Section 4.2). It will keep the pH in the range 7-10 and the redox potential at -0.30 to -0.20 V.

Steel in the waste and concrete reinforcement will corrode in water and this will be an efficient sink for oxygen left after the sealing. Degradation of organic material in the waste may also be involved in oxygen consuming reactions.

4.3.1 SFL 3

Due to the large quantities of concrete in SFL 3, the water chemistry is expected to be controlled by the leaching of the concrete. Locally and initially the pH can be as high as about 13.5 due to the alkali hydroxides in the concrete. However, with time and as a result of slow leaching and reaction the alkali hydroxides will be consumed and pH will ultimately be controlled by portlandite in concrete at about 12 to 12.5. Experiments performed and the relatively large quantities of portlandite present indicate that we will have these conditions for as long as we have to regard the long-term performance of the repository.

Locally the pH may be lower, for example in the bentonite clay mass and inside isolated waste containers. The chemistry of water inside the containers will mainly be determined by the waste and the concrete. Although the waste can be acidic, the large amount of alkalies in the concrete will neutralize the acid and keep the pH high (less than about 13.5). Presumably the redox potential is low and in the range of -0.30 to -0.15 V.

The redox conditions in this repository part will be reduced by the oxidation of the metals present such as steel reinforcement in concrete, metal containers and metal scrap in the waste. The oxygen trapped in the repository after closure will be consumed and Eh is expected to stabilise in the range of -0.30 to -0.15 V and possibly even lower than that. Despite local variations, Eh will always be sufficiently low to keep for example the important actinides in their tetravalent form once the oxygen has been consumed. The total volume of pore water inside SFL 3 will be about 5000 m^3.

4.3.2 SFL 4

SFL 4 does not contain any concrete structures, but considerable amounts of concrete and steel are present in the form of decommissioning waste. There may also be shotcrete on the walls and concrete paving of the tunnels. However, compared to SFL 3 there will be considerably less concrete. In addition to that, the waste will probably be put into steel containers which limits the contact between water and concrete. Therefore, as long as the steel containers are intact, only a moderate increase in pH is expected ($9 < pH < 12$). This is a conservative approach because a high pH will increase sorption and lower solubility. The large amounts of steel and alloys present will ensure reducing conditions to a redox potential of less than -0.15 V. The total volume of pore water inside SFL 4 will be about 11 000 m^3.

4.3.3 SFL 5

The repository contains a large concrete structure where the concrete containers are placed. Since SFL 5 is intended for core components and internal parts from the nuclear reactors, stainless steel is the major metal alloy. Other materials represented are: boron steel, boron carbide, hafnium, Zircaloy, inconel and boron glass. Corrosion of the alloys together with corrosion of concrete reinforcement will ensure a very low redox potential, possibly below -0.50 V, for as long as corrosion occurs and -0.3 to -0.15 V after that. The pH-conditions are expected to be the same as in SFL 3 due to the large quantities of concrete, i.e. 12-12.5 after an initial period of pH up to about 13.5. The total volume of pore water inside SFL 5 will be about the same as in SFL 4.

According to recent leaching experiments with OPC in both saline and non-saline waters the period with pH around 12.5 will last during at least 900 pore water exchange cycles, see Appendix C. This is in agreement with results obtained from modelling of concrete leaching by *Berner [1990]*. The number of pore water exchange cycles (900) corresponds to a time period of about 1 million years given a total flow rate through SFL 5 of about 2 m^3 annually, see Appendix C.

4.3.4 Near-field release of chemical components

Normal groundwater components such as Mg^{2+}, HCO_3^-, SO_4^{2-} and Cl^- are likely to precipitate if they are transported into the concrete filled parts of the repository. This is certain for Mg^{2+} and HCO_3^-, which will precipitate as brucite and calcite and likely for SO_4^{2-} and Cl^-. Sulphate can form ettringite in cement and chloride can at least in high concentrations form the so called Friedel's salt. However, a groundwater flow through or past the concrete filled parts can of course also pick up ions from dissolution of for example alkali hydroxides and portlandite. At least part of the near-field can therefore be expected to obtain chemical conditions that are in between deep groundwater and concrete porewater.

4.4 Data used in the prestudy

A summary of representative physical properties of barrier materials that can be found in the repository has been compiled from different information sources and is presented in Table 4.4. The materials are concrete in structures, porous concrete and gravel used as backfilling material and bentonite and sand/bentonite surrounding the concrete structures in SFL 3.

Table 4.4 Physical properties of barrier materials in the repository.

Concrete	**Porosity**	*Ref*	**Effective diffusivity** $[m^2 \cdot s^{-1}]$	*Ref*	**Hydraulic conductivity** $[m \cdot s^{-1}]$	*Ref*	**Solid density** $[kg \cdot m^{-3}]$	*Ref*
Concrete in structures	0.15	*b*	$3 \cdot 10^{-11}$	*b*	10^{-9}	*a*	2700	*b*
Porous concrete	0.3	*c*	$3 \cdot 10^{-10}$	*c*	10^{-7}-$2 \cdot 10^{-5}$	*c*	2700	***)
Gravel(sand)	0.3		$3 \cdot 10^{-10}$		10^{-4}		2500	
Pure bentonite	0.6*)	*a*	$10^{-12}/10^{-11}$ **)				1050	*a*
Sand/bentonite (90/10)	0.25	*a*	$10^{-12}/10^{-11}$ **)				2000	*a*
Sand/bentonite (85/15)	0.53	*d*					1800	*d*

a [*Wiborgh and Lindgren, 1987*]
b [*Allard et al. 1991*]
c [*Höglund, 1993*]
d [*Engkvist, 1994*]
*) uncompacted bentonite
**) 10^{-11} for cations and due to anion exclusion 10^{-12} for anions.
***) same value as for concrete structures assumed.

5 Repository site conditions

The long-lived radioactive waste in Sweden is intended to be disposed of deep in the Swedish bedrock. In the prestudy for SFL 3-5, the repository site conditions have been assumed to comply with geohydrological, thermal and geochemical conditions that could reasonably be expected at a chosen site in the Swedish crystalline bedrock. Therefore, the assessment of generic conditions for SFL 3-5 were based on results from comprehensive field investigations at the so called study sites, which were selected, investigated and evaluated prior to the KBS-3 study in 1983 [*KBS-3, 1983*].

5.1 Groundwater flow in the undisturbed rock

5.1.1 General

Flowing groundwater is a potential carrier of radionuclides from a repository. Therefore, it is important to establish the groundwater flow field at any particular site being considered to host a deep repository. A well characterised site is needed and typical site specific investigations comprise surface geological mapping, geophysical ground to air surveys and core drilled boreholes down to and beyond the anticipated repository level.

The study site investigations generally included up to 15 core drilled holes down to depths between 500 and 1000 m. In addition to that, a number of percussion boreholes were sunk to shallow depths of about 50-100 m mainly to further define the geometry and character of existing fracture zones.

The boreholes, and in particular the deep core drilled holes are geophysically logged, hydraulically tested and groundwater sampled for hydrochemical characterization. The geological conceptualisation of a site is obtained by integrating data from the surface and the boreholes.

The field of hydraulic conductivity and the groundwater surface, which in Sweden can be approximated with the land surface topography, defines the flow field. If there is a density gradient due to increased salinity with depth, this has to be taken into account in the modelling of the flow field.

Different approaches to modelling have been used. Most common is a deterministic approach where rock mass properties are averaged so that the host rock is divided into different hydraulic units like fracture zones and the rock mass in between. This approach is warranted by the fact that much of the flow is apparently concentrated to conductive zones in the rock. It is also a modelling method which is fairly straight forward to use.

However, recent research [*SKB 91, 1992*] points at the possibility of describing the rock mass as a stochastic continuum. This approach rests more or less entirely on a statistical treatment of borehole data and focuses on field measurements; the notation "hydraulic unit" as a result of averaging hydraulic properties over large distances is practically neglected. The approach makes use of an upscaling procedure between two measurement points in a borehole, so that the length over which rock mass properties have been determined, is somewhat expanded. This approach to modelling has the advantage to better describe the uncertainties involved and to separate variability from uncertainty.

Another approach is to model the hydraulic system as a discrete fracture network. This concept has been developed during the last years. The method neglects flow in the rock mass, and the flow is considered to take place only in a network of connected channels or fractures. The discrete fracture model is close to the physical reality and probably superior to the other approaches in this respect. However, the concept, which is very computer demanding, is in a state of development and so far it has mainly been applied to small scale problems. Therefore it is not yet available for performance assessment use.

5.1.2 Hydraulic conductivity in Swedish bedrock

In order to calculate the flow in and around the vaults of SFL 3-5 it is necessary to estimate the average hydraulic conductivity at repository depth in a feasible host rock. The flow calculations are needed in the performance assessment of the near-field as a barrier to radionuclide release.

In the deterministic approach, the hydraulic conductivity is generally conceived of as being decreasing with increasing depth, obeying a formula like $K=a \cdot z^{-b}$, where z is the depth below ground surface and the two constants a and b are site specific, which for the Kamlunge site is shown as an example in Figure 5.1. Also the hydraulic gradient decreases with increasing depth, generically formulated according to Figure 5.2.

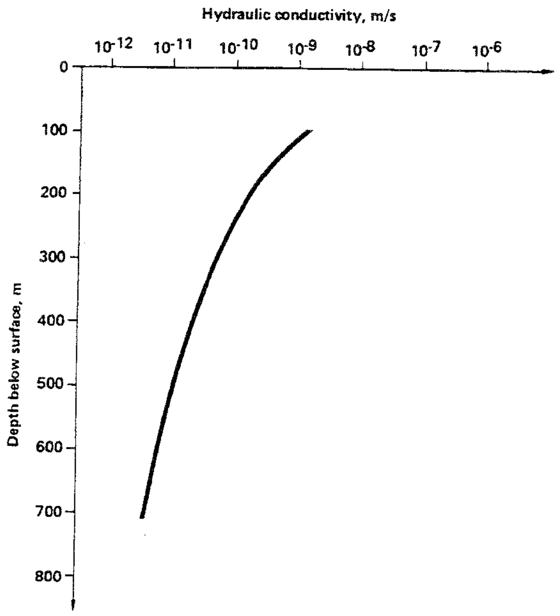

Figure 5.1 The depth dependence of the hydraulic conductivity as calculated at the Kamlunge site [KBS-3, 1983].

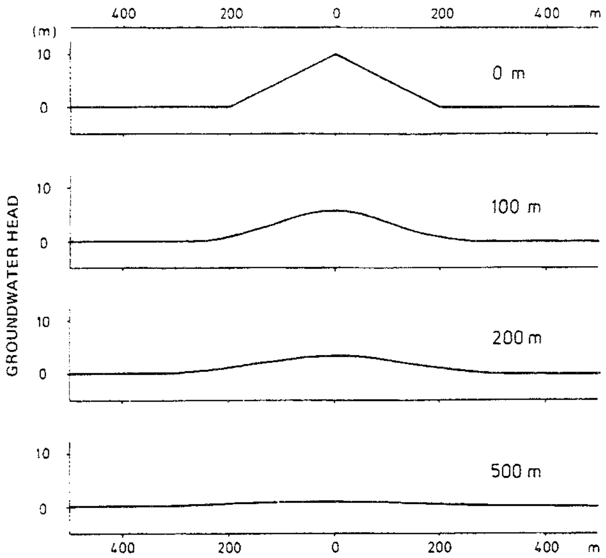

Figure 5.2 *Illustration of the levelling of the groundwater head with depth. The largest horizontal gradient at the top surface (5%) has decreased to about 0.1% at a depth of 500 m below ground surface [KBS-3, 1983].*

A number of test sites have been investigated within recent research programmes initiated by SKB, particularly within the KBS3-study [*KBS-3, 1983*]. The cited study included site investigations at the following locations: Kamlunge, Gideå, Svartboberget, and Fjällveden. Rather extensive field investigations and computer modelling of the groundwater flow at repository depth have been carried out for all four sites.

By combining the knowledge of the hydraulic conductivity and the hydraulic gradient, it is thus possible to estimate the flux at repository depth. This is done with the aid of computer modelling. From the cited report [*KBS-3, 1983*], it was found that the average flux at repository depth in the rock mass was in the range 10-100 ml/m^2,year. The higher values preferably occurred when a horizontal fracture zone is present below the repository level, but the flux values in vertical or sub-vertical fracture zones can be far higher than so. However, this can conservatively be accounted for by discarding the retention of radionuclides in fracture zones. In other words, once a radionuclide has entered a fracture zone, it is regarded as having reached the biosphere instantaneously. With this approach, possible retention in the fracture zones are not taken credit for.

A site with semi-generic data was reported in [*Lindbom et al., 1989*]. This site was modelled at three different scales with increasing degree of refinement of the grid resolution. At all three scales, the flux in the rock mass at repository depth was found to be in the range of 50-100 ml/m^2,year. The importance of horizontal fracture zones being present below repository level was pointed out also in this study. Similar to the other two studies, higher flux values were calculated in the fracture zones.

The large scale calculations flow forming part of the SKB 91-study, was based on the deterministic approach [*Lindbom et al., 1991*]. The fluxes in the rock mass at repository depth were calculated to be in the order of about 25-100 ml/m^2,year, with extremely high flux values in or at the border of the regional fracture zones circumscribing the repository area. This does, to some extent, support the strategy with the exclusion of the travel times in fracture zones (see previous discussion).

The SKB 91-project [*SKB 91, 1992*] also aimed at describing the flow conditions at a smaller scale at the Finnsjön site by applying the stochastic continuum approach. At this scale and with this method, the site appeared to have a more conductive rock mass, which resulted in somewhat higher flux values at repository depth. The bulk of calculated fluxes were in the range 100-1000 ml/m^2,year.

In conclusion, for the forthcoming calculations in the present study, it seems feasible to assume that the fluxes in the rock mass at repository depth are 100 ml/m^2,year; an assumption that seems to be in reasonable agreement with average values from the previously cited studies.

5.2 Temperature at repository depth

The SFL 3-5 repository will according to the present concept be situated at about 500 m depth. The temperature increases with increasing depth. Measurements in bore holes down to about 500 m level at the Finnsjön site indicate a temperature gradient of 1.3 °C/100 m and the temperature at 500 m level was found to be 12.3 °C. From two series of measurements in the borehole Gravberg-1 at Siljan, the temperature gradient was found to be 1.6 °C/100 m [*Birgersson et al., 1992*].

5.3 Groundwater composition

Groundwater at the site will infiltrate and fill the vaults after the repository has been sealed and closed. Interactions of water and water constituents with the construction and backfill materials, and with the waste have to be accounted for in the performance assessment. Influence on radionuclides are particularly important in this context. It is therefore necessary to know the composition of deep groundwater, the possible variations in composition and the influences from materials in the repository on the composition.

Groundwater under land in Sweden has in general a meteoric origin. Even at sites were saline waters were encountered, a large portion of meteoric water was indicated by the isotopic composition of the water [*Ahlbom and Smellie, 1992*]. Infiltrating meteoric water contains oxygen, but this is largely consumed in the soil and further along the flow path in the rock. Biological processes mainly in the soil uses the oxygen and generates carbon dioxide which reacts with minerals like for example calcite and an input alkalinity is obtained. Further geochemical reactions in the rock groundwater system influence the composition, redox properties and pH of the water. This includes reactions with iron and sulphide minerals, organic materials, weathering of feldspars, ion exchange in clay minerals etc. It has been demonstrated that the iron system defines a redox potential with Eh values of about -0.3 to -0.1 V. The pH is generally buffered by the carbonate system. An alkali input beyond the control of the bicarbonate content of the groundwater will ultimately be limited by reactions with the silicate minerals which become reactive at a pH of about 9 to 10, but this is presumably a relatively slow reaction.

Saline groundwater with chloride as the dominating anion, mainly balanced by sodium and calcium, is sometimes encountered at locations near the sea shore, places where Sweden have been covered by the sea prior to land rise, or at great depths. Different origins may be envisaged. However, the point is that we have to consider the possibility of saline groundwater, at least until a site have been selected and investigated. Saline water is not a hindrance to the location of the repository. The SFR facility under the sea floor in Forsmark is a good example of that.

In cases where sea water has infiltrated it can frequently be seen that sulphate is reduced and bicarbonate concentration increased. However, chloride anions only react to a minor extent with the minerals and chloride is therefore regarded as a non-interacting (conservative) groundwater constituent. The ratio between Na^+ and Ca^{2+} can vary which is probably a result of reactions with the minerals such as weathering, precipitation and ion exchange. The concentration of K^+ and Mg^{2+} is suppressed by geochemical reactions and controlled at a relatively low level. Fluorine is controlled by the solubility of calcium fluoride (fluorite).

The main constituents in groundwater and their typical concentration ranges are listed in Table 5.1.

Table 5.1 Ranges of groundwater composition. Concentrations in mg/l.

Species	**Expected conc. in non-saline waters**[a]	**Measured conc. in saline water from Finnsjön area**[b]
pH	7-9	7.5-8.8
Na^+	10-100	875-1500
K^+	1-5	7-10
Ca^{2+}	10-40	630-1790
Mg^+	1-10	70-140
HCO_3^-	90-275	30-170
SO_4^{2-}	0.5-15	200-320
Cl^-	4-15	2600-5600
F^-	0.5-5	0.7-2
Fe_{tot}	1-5	2-3
Mn	0.01-0.5	0.4-0.8
HS^-	0-0.5	
NH_4^+	0.05-0.2	
NO_3^-	0.01-0.5	
NO_2^-		
$PO_{4,\,tot}$	0.01-0.2	0.05-0.4
TOC	1-8	

a [*KBS-3, 1983*]

b Borehole Fi5, 141-384 m, [*Laurent, 1982*]

When groundwater infiltrates the vaults after closure and sealing, there will be a strong chemical influence on the composition. For example concrete porewater has a high pH and is rich in ions such as Na^+; K^+ and Ca^{2+}. This is further discussed in Chapter 4. The large amounts of concrete will influence the chemical conditions inside the vaults and there may also be some increase of pH and Ca^{2+}-concentration in the near-field. There will also be an initial release of Na^+ and K^+. Observations in the Äspö Hard Rock Laboratory indicates that especially potassium may increase in concentration above ambient in the near-field as a result of concrete injection [*Wikberg, 1994*]. However, no attempt have been made in this study to estimate the magnitude of the chemical influence from concrete.

5.4 Data used in the prestudy

In the prestudy the repository site conditions have been assumed to be representative for typical Swedish bedrock.

The hydraulic situation at the site is assumed to be constant with time and is given by a water flux of 0.1 l/m^2,year in the undisturbed rock. The temperature at repository depth is assumed to be approximately 10 °C. The groundwater in the surrounding rock is assumed to be reducing and with a pH of about 8. It is also assumed that both saline and non-saline groundwaters can be found at the repository depth.

6 Scenario selection

6.1 General

Safety assessments of radioactive waste repositories are based on predictive modelling of the performance of engineered barriers and natural barriers for very long time scales. To evaluate the performance of a repository, assumptions must be made on the future evolution of engineered barriers and natural conditions. Formulation of scenarios is a way to describe possible sequences of processes and events influencing the release of contaminants from a repository.

Early performance assessment studies have used so called "Judgemental methods" to identify and formulate scenarios. However, the need for more systematic scenario development methods has been identified, and such methods have recently, or are presently tested in Sweden and elsewhere. One method developed by the Swedish Nuclear Power Inspectorate, SKI, in the SITE-94 project is based on Influence Diagrams with linked documentation [*SKI, 1995*]. A preliminary report on this work is found in [*Chapman et al., 1994*]. The methodology builds to a large extent on the results of the Joint SKI/SKB Scenario Development Project [*Andersson, 1989*]. In this joint scenario exercise Features, Events and Processes, FEPs, considered relevant for deep disposal of spent fuel in granitic rock were compiled, described, sorted and screened. The majority of the FEPs remaining after screening were assigned to the Process System, PS, which were defined as:

> " the organised assembly of all phenomena (FEPs) required for the description of barrier performance and radionuclide behaviour in a repository and its environment, and that can be predicted with at least some degree of determinism from a given set of external conditions."

FEPs kept outside the Process System, External FEPs, were considered as scenario generating FEPs, and hence scenarios are formed by combining the Process System with one or a combination of External FEPs.

To be able to evaluate how the Process System responds to External FEPs a structuring of the Process System is needed. One way to do this is to construct an Influence Diagram of the PS where FEPs within PS are represented by boxes and interactions between FEPs are illustrated by lines between the boxes.

The above methodology developed by SKI has in the present study been applied by SKB on the SFL 3-5 concept. This Chapter gives an overview of the application, and the results in terms of a Reference Scenario for the long-term behaviour of the different repository parts. A more detailed description can be found in [*Skagius and Wiborgh, 1994*]. The terminology used are defined in Appendix A.

6.2 Methodology

The methodology with Influence Diagrams involves the following main steps:

- Construction of a Basic Influence Diagram
- Development of Scenario Influence Diagrams from the Basic Influence Diagram
- Formulation of scenarios and calculation cases.

6.2.1 Construction of Basic Influence Diagram

The Basic Influence Diagram should ideally contain all FEPs which are relevant for the system studied for any scenario. This requires a definition of the system in terms of waste form, barrier design and materials as well as repository lay-out. In addition, decisions must be taken on the geometrical extension of the system to be included in the Influence Diagram, i.e. the extension of the Process System. Ideally, the Process System should comprise the whole universe and be described in greatest possible detail, but this is of course not possible. The aim of the assessment will have to determine where the system boundary is set and to what level of detail the repository components inside the system boundary should be described in the Influence Diagram.

The Process System for SFL 3-5 has in the prestudy been restricted to include only the engineered barrier system and the nearby rock disturbed by the excavation and the presence of the repository. Due to the difference in both the waste and the engineered barrier system between the three repository parts the Influence Diagram was divided into four main regions, one for each repository part and one for the near-field rock. The regions representing the repository parts were further divided into sub-regions representing the different barriers, as illustrated in Figure 6.1.

Existing FEPs lists for radioactive waste disposal [*Andersson, 1989, Stenhouse et al., 1993*] and experiences from earlier investigations of SFR formed the basis for selection of FEPs relevant for the defined Process System. Each selected FEP was then introduced into the Influence Diagram as a box with the FEP name in each sub-region of the diagram representing a barrier where the FEP could occur. Within a sub-region, FEP-boxes were ordered such that phenomena directly involved in the transport of radionuclides and chemotoxic elements were represented at the bottom of the sub-region and FEPs affecting barrier properties at the top. The physical and chemical conditions in the barriers constitute the link between these two groups of FEPs, see Figure 6.2.

After all known FEPs have been compiled and introduced into the Influence Diagram, interactions between FEPs are identified and indicated in the diagram. This is done by drawing a line between the interacting FEPs, and with an arrow showing the direction of the influence. Each influence in the diagram is marked with a unique code. To facilitate the evaluation of the Influence Diagram it is important that an interaction is described as an influence on the primary target FEP. There are no restrictions on the number of influences between two FEPs, since one FEP may influence another FEP in several, different ways.

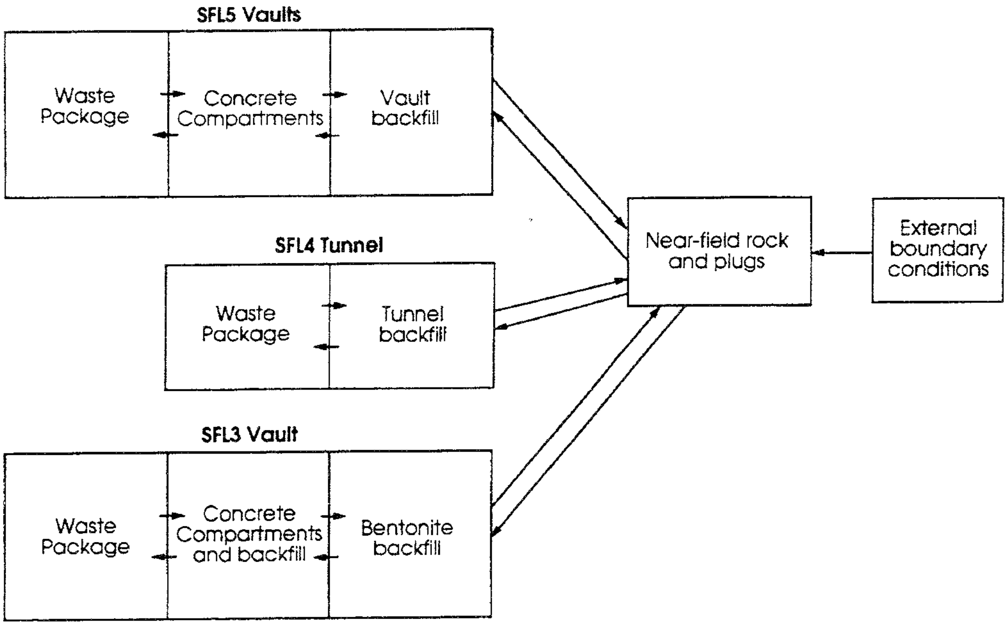

Figure 6.1 Schematic lay-out of the Influence Diagram for SFL 3-5.

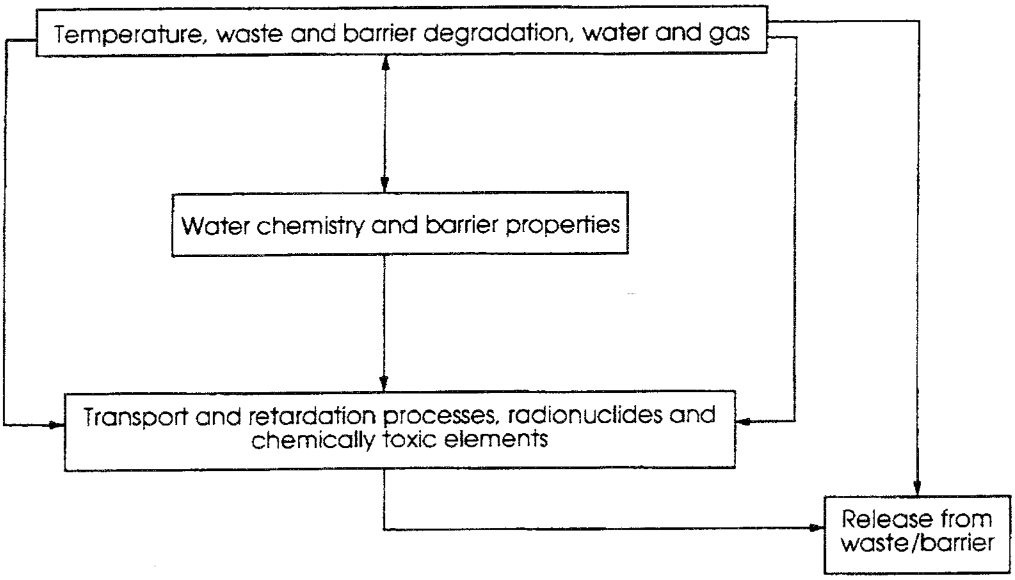

Figure 6.2 Schematic lay-out of how the FEPs are assigned to different areas in the Influence Diagram for SFL 3-5.

To avoid confusion and misinterpretations of which phenomenon a FEP-name in the Influence Diagram is referring to, documents describing the FEPs were prepared. These documents are stored in a database, and each FEP-box in the Influence Diagram is coupled to the corresponding FEP-document in the database. Similarly, all influences in the diagram are coupled to documents containing the influence code, the names of the FEPs between which the interaction occurs, a specification of the interaction and references to literature, if available.

6.2.2 Development of Scenario Influence Diagrams

The Process System in the Basic Influence Diagram contains FEPs and influences which may affect the behaviour of the repository system, but at this stage no evaluation of the significance of the different FEPs and influences on the repository performance has been made. Since the significance of FEPs and influences depends on the initial conditions of the Process System and how the Process System is affected by the surroundings, the next step is to define these entities, i.e. to define the premises of the scenarios to be evaluated. Once this is done, the Influence Diagram is reviewed and the significance of each influence is assessed for the premises of a specific scenario. This results in an Influence Diagram for this specific scenario.

Since there is a large number of FEPs and influences that should be reviewed and judged, the evaluation of an Influence Diagram for a selected scenario is time consuming. By defining a simple Reference Scenario and make a complete evaluation of the Influence Diagram for the Reference Scenario, time can be saved since the evaluation of additional, more complex, scenarios can be restricted to parts of the Influence Diagram that differs from the Reference Scenario diagram.

The procedures for developing a Reference Scenario Influence Diagram and Influence Diagrams for other scenarios are described below.

Reference Scenario

The premises for the Reference Scenario selected for SFL 3-5 are as follows:

- the repository is closed and the properties of the barriers are in accordance with design criteria,
- the hydrological, hydrochemical, rock mechanical and thermal conditions at the Process System boundary, i.e. at the interface between near-field and far-field rock, are representative for typical Swedish bedrock and constant with time.

An Influence Diagram for this Reference Scenario was developed by assessing the significance of each influence in the Basic Influence Diagram for the above specified conditions. At the same time the Influence Diagram was reviewed in order to improve the description of the Process System. Ideally, the significance assessment and the review should be carried out by a number of people with expertise in different disciplines in order to cover all aspects of the long-term performance of an underground repository. This was not possible to achieve within this study. However, the full diagram was reviewed by at least two persons, and part of the diagram was reviewed by the full Project Group involved in the prestudy.

Before assessing the significance of influences an assessment scale has to be selected and each level of the scale has to be defined. Here, a three-level significance scale was used. Influences judged to be insignificant for the target FEP were labelled "negligible", influences which were assessed to be significant for the target FEP were labelled "important", and influences which presently could not be judged either as negligible or important were labelled "uncertain".

The assessed significance together with the reasons behind the assessment were documented in a protocol coupled to each influence in the Influence Diagram. A motivation for the significance is especially important for those influences assessed to be negligible since they will not be considered in the subsequent evaluation of the defined scenario. Ideas on how to consider the influence in modelling were also documented in the protocol. In addition, the people involved in the significance assessment and review and their degree of expertise in the topic were documented in the protocol.

When the significance of all influences in the diagram was assessed for the Reference Scenario premises, a first version of the Influence Diagram for the Reference Scenario was completed. The next step is to reduce the Influence Diagram to a desired significance level. Because of the coarse significance scale used it seemed appropriate at this stage to remove only influences assessed to be negligible. The unreduced Reference Scenario Influence Diagram contains approximately 1200 influences and about 900 of these remain in the reduced Influence Diagram. Approximately 30% of the influences in the reduced Influence Diagram was assessed as "uncertain". The quite large number of influences still remaining in the reduced diagram could seem a bit disappointing, but it should be pointed out, that this is partly due to limitations in disciplines covered by the people involved in the significance assessment. A larger and more appropriate composed assessment group would probably have provided arguments for assessing a larger number of influences as negligible already at this stage.

Other scenarios

By using the Influence Diagram for the Reference Scenario, Influence Diagrams for other scenarios can be developed. It has not been possible to test this part of the methodology within the frame of the prestudy. Therefore only a general description of the procedure is given below.

As stated in the beginning of this Chapter, a scenario is formed by combining the Process System with one or a combination of External (scenario initiating) FEPs. Hence, the External FEPs corresponding to the premises of the scenarios to be studied must be defined. For example, External FEPs for a scenario for climatic evolution could be "Sea-level changes", "Permafrost" and "Glaciation" [*Chapman et al., 1994*]. To evaluate the impact of the selected External FEPs on the Process System, the primary target FEPs in the **unreduced** Influence Diagram for the Reference Scenario is first identified. The significance of influences down-stream of these primary target FEPs are then re-evaluated considering the potential disturbance caused by the attack of the External FEP. If the significance of an influence is assessed to be the same as in the Reference Scenario further evaluation of influences and FEPs downstream along this route is not needed, unless the nature of the influence is different from that in the Reference scenario. In this way an Influence Diagram for the selected scenario is developed where the difference between the selected scenario and the Reference scenario is given by a few paths in the diagram.

In the same way as for the Reference scenario the evaluation of other scenarios should be documented in protocol linked to each influence in the Influence Diagram. Reduced Influence Diagrams at different significance levels are prepared by removing influences and FEPs.

The selection of scenarios is a difficult task since there is a large number of External FEPs and combination of External FEPs that can affect the Process System. It is believed that the number of scenarios to be evaluated will remain large, unless a judgement of the most probable/critical External FEP or combination of External FEPs is made a priori. Alternatively, the most critical pathway through the Process System could be identified without specifying how this pathway is activated, i.e. totally disregarding External FEPs. It should be pointed out that one can never guarantee that the description of the Process System is complete. However, the uncertainty in the description of the Process System will probably decrease as the number of reviews and evaluations of the Process System increase. This suggests that at least a couple of scenarios defined by External FEPs should be evaluated prior to identifying a critical pathway through the Process System.

6.2.3 Formulation of scenarios and calculation cases

The reduced Reference Scenario Influence Diagram was used to formulate the Reference Scenario and identify calculation and modelling needs for quantitative estimates of the release of radionuclides and chemotoxic elements from the repository. Since this work and subsequent calculations were carried out as a part of a prestudy of the SFL 3-5 repository concept it was not possible to consider all phenomena and influences remaining in the reduced Influence Diagram. Therefore a Reference Case for the Reference Scenario was formulated and analysed in the prestudy. This Reference Case as well as phenomena remaining to be addressed in a full Reference Scenario analysis are described in the following Section 6.3.

Also at this stage the documentation is essential. For this purpose, the protocols linked to the Influence Diagram are used. The documentation should include a description of how influences and FEPs are considered in the models and the assessment calculations as well as the results of the calculations and associated uncertainties. This documentation will be valuable in forthcoming studies.

The outcome of the performed studies and calculations will increase the understanding of the behaviour of the system. The Influence Diagram should therefore be revisited and updated based on increased knowledge of the significance of interactions between FEPs.

6.3 Reference Scenario for SFL 3-5

The reduced Influence Diagram for the Reference Scenario was used to identify phenomena and aspects of phenomena that are of potential importance for the long-term performance of the SFL 3-5 repository and to select calculation cases for the prestudy. In this section the processes and mechanisms considered in the analyses within the prestudy are described and phenomena of potential importance, but not addressed in the prestudy, are listed.

6.3.1 Description of selected Reference Case

The Reference Case selected for analysis is described following the migration of radionuclides and chemotoxic elements through the near-field barriers from mobilisation to release. The Reference Scenario boundary conditions stating constant hydraulic situation and stable groundwater chemistry are applied. After repository closure groundwater in the far-field rock will resaturate the near-field rock and engineered barriers in the repository. The hydrological and chemical conditions during the resaturation phase will probably differ from the conditions established once the repository becomes fully resaturated. However, the duration of the resaturation phase is short in relation to the time perspective considered and is therefore not included in the selected Reference Case. Consequently, the initial conditions assumed are that all near-field barriers are fully resaturated and reducing conditions have been re-established. Furthermore, the water has entered the waste packages and are in contact with the waste. Potential interactions between the different repository parts are not included in the calculation cases.

SFL 3

The waste allocated to SFL 3 is a mixture of various materials. Radionuclides and chemotoxic elements in the waste are assumed to immediately dissolve in the water in the waste packages. The concentration of the elements in the water is limited by solubilities and sorption on cement and concrete in waste and packaging. Slow corrosion of metal waste may further limit the concentration of radionuclides present as induced activity in the metals and any chemotoxic elements included in the metals. In addition, the presence of stable isotopes may affect the concentration of radioactive isotopes of the same element (isotopic dilution).

The solubility and extent of sorption are dependent on the water chemistry in the waste packages. The presence of cement and concrete will establish a high pH and ionic strength, and corrosion of steel waste and packaging as well as steel reinforcement in concrete packaging will maintain reducing conditions in the packages. The water may also contain organic complexing agents as a consequence of decomposition of organics in the waste in the alkaline water in the packages.

All cement and concrete surfaces in the waste and packaging are assumed to be immediately accessible for sorption, but the waste packaging materials provide no transport resistance for dissolved radionuclides and chemotoxic elements.

Dissolved radionuclides and chemotoxic elements in the waste packages are assumed to be transported through surrounding porous concrete backfill and concrete compartments by diffusion in interconnected pores and fractures, which are assumed to be evenly distributed in the materials. The concentration of these elements in the water in the compartments is limited by the solubility and by sorption on concrete backfill and compartment structures. Likewise to the waste packages, the pH and ionic strength of the water in the concrete compartments are high and reducing conditions prevail. In addition, the water is assumed to contain organic complexing agents formed by alkaline decomposition of organics in the waste package and/or in the concrete compartments.

Further out-transport of radionuclides and chemotoxic elements through the surrounding clay barriers is assumed to be dominated by diffusion. The concentration of the elements in the pore water is limited by the solubility and by sorption on the clay materials. Here, the water chemistry in terms of pH, ionic strength and redox conditions is assumed to be determined by the clay materials in contact with reducing groundwater. Radionuclides and chemotoxic elements which exist as anionic species in this kind of water will have a smaller accessible diffusion porosity than other species in the water due to anion exclusion.

Radionuclides and chemotoxic elements which have penetrated the clay barriers are by diffusion transferred to the groundwater flowing in the near-field rock and then moving with the groundwater through the near-field rock without any interaction with the rock minerals. Due to disturbances in the near-field rock during repository excavation, the hydraulic conductivity and thus the water flux in the near-field rock is higher than in the far-field rock.

SFL 4

The waste materials to be disposed in SFL 4 are metals and concrete. Radionuclides and chemotoxic elements in the waste are assumed to be immediately dissolved in the water in the waste packages. The concentrations of these elements are limited by their solubility in the water, but potential effects of sorption on concrete waste and metal materials are disregarded. Some of the metal waste contains induced activity and the concentration of these radionuclides as well as of any chemotoxic elements included in the metal may be further limited by slow corrosion of the metals. The presence of both stable and radioactive isotopes of the same element may lower the concentration of radioactive isotopes (isotopic dilution).

Corrosion of steel packaging and metal waste will maintain a low redox potential in the water in the packages and the presence of concrete waste may create an alkaline environment.

The steel containers are assumed to be permeable enough to allow water flowing in the surrounding sand/gravel backfill to also flow through the containers. Dissolved radionuclides and chemotoxic elements leaving the containers with the flowing water are further transported with the moving water and by diffusion through the sand/gravel backfill in the tunnel and through the near-field rock. The backfill is assumed to be homogeneous and significantly more permeable than both the near-field and far-field rock. Consequently, the hydraulic conductivity of the backfill and the hydraulic situation in the far-field rock will determine the water flux through the tunnel backfill and near-field rock.

The release of radionuclides and chemotoxic elements to the far-field rock is delayed due to sorption on the backfill material. Any interactions between these elements and the near-field rock are disregarded. Out-transport of alkali and calcium hydroxides leached from the concrete waste might increase the pH and ionic strength of the water in the backfill and thereby affect the speciation and sorption of radionuclides and chemotoxic elements on the backfill material.

SFL 5

SFL 5 is the repository part intended for reactor internals, i.e. metal components with both induced activity and surface contamination. Radionuclides deposited on the surface of the metal components are assumed to be immediately dissolved in the water in the waste packages. Activation products and chemotoxic metal elements are liberated and dissolved as corrosion of the metal components proceeds. Solubilities and sorption on concrete backfill in the packages will, however, limit the concentration of dissolved radionuclides and chemotoxic elements in the package interior. In addition, the presence of stable isotopes will influence the concentration of radioactive isotopes of the same element in the water.

Dissolved radionuclides and chemotoxic elements are transported by diffusion and water flow through pores and fractures in the concrete packaging and surrounding compartment structures, but the release will be delayed due to sorption on the concrete materials. Initially, the permeability of the concrete compartment structures and packaging is significantly lower than the permeability of the sand/gravel backfill surrounding the compartments. Groundwater entering the vault will then mainly flow through the sand/gravel backfill without passing through the concrete compartments. In the long-term perspective, leaching of the concrete materials may increase the porosity of the concrete barriers which could affect both the hydraulic situation and diffusion flux through the barriers as well as the amount of concrete surfaces available for sorption.

Solubilities and sorption of radionuclides and chemotoxic elements in concrete waste packages and compartments are assumed to be determined by high pH and ionic strength and a low redox potential due to the presence of concrete and corroding steel waste and reinforcement in concrete packaging and compartment structures. Long-term changes in pH and ionic strength may occur as a result of leaching of alkali and calcium hydroxides from the concrete materials.

Radionuclides and chemotoxic elements released from the concrete compartments by diffusion and moving water will be captured by the groundwater flowing around the compartments in the sand/gravel backfill in the vault. This water flow, and diffusion, will transport dissolved radionuclides and chemotoxic elements through the backfill and further out through the near-field rock. Likewise to the SFL 4 tunnel, the high permeability of the backfill in the SFL 5 vault will together with the hydraulic situation in the far-field rock determine the water flow through the backfill and the near-field rock.

Sorption on the backfill material will delay the release of radionuclides and chemotoxic elements from the near-field rock surrounding the SFL 5 vault. Leaching of concrete compartment structures and packages might create an alkaline environment in the sand/gravel backfill which affect the speciation and sorption on the backfill material.

6.3.2 Remaining issues

There are a number of processes and mechanisms which have been assessed to be of potential importance for the long-term performance of the repository, but which so far have not been considered in the analyses of the Reference Scenario. Future studies should include an assessment of the potential for these processes and mechanisms to occur at expected conditions in the repository as well as of the impact on the release of radionuclides and chemotoxic elements from the repository.

Radionuclide migration processes

Processes directly involved in the mobilisation and out-transport of radionuclides and chemotoxic elements which so far have not been considered in the calculations are:

- gas generation and transport,
- sorption/coprecipitation with corrosion products,
- coprecipitation with calcite, and
- colloid generation and transport.

Gas will be generated in the repository, mainly due to corrosion of metals. The impact of gas generation and gas flow on saturation conditions and water flow as well as the release of radionuclides and chemotoxic elements in gaseous phase have not been assessed in the prestudy.

There are indications that iron corrosion products act as a scavenger for trace elements in solution. Sorption/coprecipitation with corrosion products could then be an important retardation mechanism for radionuclides and chemotoxic elements since the repository contains a substantial amount of corroding steel.

Coprecipitation with calcite may be an important retardation mechanisms for radionuclides and chemotoxic elements which form carbonate minerals. This could be worthwhile to include in future assessments.

Colloids can act as carriers for radionuclides and chemotoxic elements. The possibility of forming colloids in the repository and their mobility in the repository barriers should therefore be estimated.

Barrier alteration/degradation processes

The different near-field barriers in the proposed repository design may in the long-term perspective be altered and even degraded due to interaction and chemical reaction with the water and its constituents and due to mechanical impact. The only alteration/degradation process considered in the prestudy analyses of the Reference Scenario is leaching of cementitious components from the concrete barriers in SFL 5. Other processes and mechanisms presently assessed to be of potential importance for the long-term properties of the near-field barriers are listed below.

- Leaching of the concrete barriers in SFL 3; effects on chemical conditions, may increase the permeability to water and gas and reduce the mechanical stability of the barriers.

- Precipitation of cementitious components in near-field barriers; may decrease the permeability to water and gas.

- Corrosion of iron reinforcement and precipitation of volume expanding corrosion products in concrete barriers; effects on mechanical stability and permeability to water and gas.

- Formation of volume expanding ettringite/monosulphate or Friedel salts (chlorides) in concrete barriers; effects on mechanical stability and permeability to water and gas.

- Gas pressure build-up in concrete barriers; effects on mechanical stability and permeability to water and gas.

- External forces on (altered) concrete barriers; mechanical load, bentonite swelling pressure, volume expansion of adjacent barriers may mechanically impact concrete barriers affecting the permeability to water and gas.

- Microbes in concrete barriers; effects of biofilms on sorption properties and effects of clogging on permeability to water and gas.

- Corrosion of steel packaging; effects on permeability, sorption properties and mechanical stability of packaging.

- Bentonite swelling; effects of expansion of high density bentonite backfill into low density sand/bentonite backfill on swelling pressure and hydraulic properties of the clay barriers.

- Chemical alteration of clay barriers; effects of interaction with cementitious and waste components on swelling capacity and plasticity and thereby hydraulic and transport properties of the clay barriers.

- Mechanical impact on (altered) clay barriers; effects of rock movements on transport properties.

- Dilution of clay barriers and sand/gravel backfill; potential for and effects on hydraulic properties of increased volume available for the clay barriers and sand/gravel backfill due to leaching of inner concrete barriers and out-transport of dissolved components.

- Precipitation of cementitious components and corrosion products in sand/gravel backfill; effects on permeability to water and gas.

- Geochemical alteration of near-field rock; effects of precipitation of or chemical reaction with components dissolved from engineered barriers and waste on permeability to water and gas and on geochemical processes affecting contaminant transport, e.g. sorption.

- Mechanical alteration of near-field rock; potential for fracturing of near-field rock due to creep, gas pressure build-up in the repository vaults and loss of mechanical support as a consequence of volume decrease of engineered barriers, and effects on the flow situation in the near-field rock.

- Alteration/degradation of vault and shaft seals; potential for and effects on flow situation in the repository area.

Barrier alteration/degradation and migration processes are to a large extent dependent on the evolution of the water chemistry in the repository barriers, but they may also affect the water chemistry. Concrete leaching and steel corrosion are important in this sense and effects on pH and redox conditions from these processes are considered in the Reference Case. Another impact on water chemistry which is not included in the Reference Case is the dissolution of the waste in SFL 3. For example, ion-exchange resins may degrade and thereby liberate sulphates and amines. Sulphates may cause ettringite formation in concrete barriers, and amines may form complexes with radionuclides and chemotoxic elements. Furthermore, some of the waste types contains ferrocyanide precipitates which if dissolved may influence the solubility and sorption of radionuclides and chemotoxic elements by the formation of cyanide complexes.

The resaturation phase is not considered in the Reference Case. Despite the short duration of the resaturation phase in relation to the time of interest of repository performance, there are some aspects of the resaturation phase that may be of importance. Unsaturated conditions during the resaturation phase may delay the initiation of barrier and waste alteration/degradation processes, such as corrosion of steel packaging and metal waste and dissolution of other waste types. This time delay may affect the release of short-lived radionuclides from the repository. Furthermore, the presence of entrapped air in the repository and the dissolution of the air in the intruding water will initially affect the chemistry of the water, e.g. redox conditions by dissolved oxygen. This may initiate barrier alteration processes which are different from those active in a long-term perspective.

7 Chemistry of radionuclides and toxic elements in the repository

The chemical conditions in the repository will influence the mobility of elements in the waste as well as the release of elements from the repository. The concentrations of many elements are limited by sorption in the barrier materials. In addition, the concentrations of some elements are limited also by their solubility inside the waste packages. The solubility and sorption data are presented in this chapter for the different chemical conditions in the repository barriers e.g. concrete, bentonite, sand/bentonite mixtures and sand backfill. The values given are best estimates. Mechanisms and processes that may have an influence on the availability of the elements in the waste and the subsequent release are discussed. These are:

- co-precipitation with and adsorption on corrosion products
- content of inactive species and potential influences
- content of organic material and formation of complexing agents
- colloid generation and their effect on radionuclide transport.

7.1 Sorption in barrier materials

The extent of sorption in different barrier materials depends on the water chemistry, the material composition and accessible material surfaces. The aim is to derive a realistic data set for radionuclides of with respect to safety for sorption in the different barrier materials present in the cement based repository. As in all databases, the selection should be justified. This is, however, difficult since values published on sorption do not always agree. The values published are often "single values" where the pH dependence, ionic strength dependence and influence of complexing agents were not studied. This is of course a result of the difficulties of changing the pH and ionic strength in a cementitious system which is highly buffered by the dissolution of alkali hydroxides and the leaching of portlandite. However, with knowledge of the sorption mechanisms and the chemistry of the studied element, it is possible to make predictions of the sorption behaviour and check the relevance of measured data.

The selection of sorption data for this prestudy is based on published values [*Allard et al., 1991, Bradbury and Sarott, 1994, NAGRA, 1993, Albinsson, 1991*] and some new experimental values [*Engkvist, 1994*], see also Appendix C. Expert judgement was used in a few cases where experimental values were not available for example the sorption properties of Cd and Be.

7.1.1 Sorption in concrete materials and sand

The difference in sorption of important radionuclides on sand and degraded concrete is considered to be negligible if the values used for sorption on sand correspond to high ionic strength or elevated pH (>9). This has been demonstrated by experiments. Radionuclide sorption experiments have been performed on different cements and cement mixtures including Ordinary Swedish Portland cement (Degerhamn anläggningscement). It has been found that the differences in sorption are of minor importance [*Allard et al.,* *1984*]. However, the ballast material plays a significant role in the sorption of the radionuclides. The mineral composition of the ballast is not critical within the range of commonly used rock types, but it is the solution phase and in particular its pH that is of most importance for sorption behaviour. The only exception is when radionuclides sorb by ion exchange. This is typical for monovalent ions and some divalent, eg. cesium, strontium and radium where the mineral composition can influence the sorption significantly [*Albinsson, 1991*].

The pH of concrete porewater and concrete leachates is very high (about 13.5). If the alkali hydroxides are leached away, the pH drops to about 12.5, buffered by the solubility of portlandite. It has been argued that the tetravalent actinides form negatively charged hydroxide species ($An(OH)_5^-$) at pH 13.5, which would possibly lower the sorption of the actinide. However, recent studies have shown that such anionic species are not formed [*Engkvist, 1993*] and therefore the high pH is not affecting the sorption properties negatively.

The sorption properties of di- and trivalent elements (with the exclusion of Sr) can be regarded constant in the interval $12.5 < pH < 13.5$, see Table 7.1.

The relatively high ionic strength in the water in a concrete containing repository, lowers the sorption of the monovalent elements which are sorbed by ion exchange. The sorption of these elements is expected to increase if the concrete is leached.

Experiments with porous concrete from the silo in SFR show that the sorption is not dependent on w/c [*Engkvist, 1994*]. This implies that no new phases are formed at high w/c or alternatively, that those formed get the same sorption properties as the ones formed at low w/c. The migration rate is strongly dependent on the w/c but this is only an effect of the increasing porosity with increasing w/c. The sorption data set in Table 7.1 are valid for "ordinary" concrete (w/c=0.5) and for porous concrete (w/c=1.4).

Cesium

Cesium ions are not hydrolysed in water. Therefore the sorption of cesium is independent of the pH. However, cesium will compete with calcium and sodium present in fresh concrete porewater and yield a K_d-value of about 0.002 m^3/kg for cesium sorption in fresh concrete. Sorption will increase if the concrete is leached and the concentration of competing cations decreases. When the concrete has been leached, the distribution coefficient for cesium will increase to 0.1 m^3/kg [*Allard et al., 1991*]. It should be noted that for cesium the ballast material and not the cement paste in the concrete determines the magnitude of sorption [*Andersson and Torstenfelt, 1983*]. For the sorption of cesium on sand the sorption of cesium on granite at high ionic strength [*Albinsson, 1991*] has been selected as representative.

Strontium, radium, cobalt, nickel, iron, beryllium, cadmium and lead

Sorption of the divalent elements is generally rather low. These metal ions are weakly hydrolysed but form anionic hydroxide species in highly alkaline medium. The sorption property of strontium is related to the concentration of calcium as a competitive ion and is expected to increase if the concrete becomes leached. A K_d-value of 0.002 m^3/kg is chosen for fresh and 0.02 m^3/kg for leached and degraded concrete [*Allard et al., 1991*]. For the sorption on sand, the K_d-value 0.01 m^3/kg [*Albinsson, 1991*] is chosen to be representative.

There is not much information about the sorption of radium on cementitious material in the literature. The K_d-value of 0.05 m^3/kg has been taken from NTB 93-08 for fresh, leached and degraded concrete [*Bradbury and Sarott, 1994*]. For sand, the same K_d-value 0.1 m^3/kg as for sorption in granite at high salinity (high ionic strength) has been selected to be representative [*Albinsson, 1991*].

Sorption of nickel on fresh and leached concrete is considerable. Nickel sorbs onto the ballast material as well as onto the CSH-gel. The K_d-value 0.1 m^3/kg has been selected as representative. This a lower value than reported previously (e.g. SFR 91-02, 1 m^3/kg). However, a critical review of earlier experiments and new accurate sorption studies of nickel at very low nickel concentrations support the lower value 0.1 m^3/kg [*Pilkington and Stone, 1990, Bradbury and Sarott, 1994, Engkvist 1994*]. For the sorption in degraded concrete as well as on sand, the K_d-value 0.02 m^3/kg [*Albinsson, 1991*] is chosen to be representative.

There are few measurements on sorption of cobalt and iron on cementitious material. However, the sorption behaviour of cobalt and iron should be similar to nickel. Therefore the K_d-value 0.1 m^3/kg has been selected for fresh and leached concrete. This is supported by recent experiments with cobalt at Chalmers [*Engkvist, 1994*]. In analogy with nickel, a K_d-value of 0.02 m^3/kg is chosen for sorption of cobalt and iron in degraded concrete and on sand [*Albinsson, 1991*].

Beryllium and cadmium are expected to behave similarly to radium and K_d=0.05 m^3/kg has been selected to represent sorption of beryllium and cadmium on fresh, leached and degraded concrete. A higher value for sorption into sand due to a lower ionic strength compared to cementitious conditions is expected and K_d=0.1 m^3/kg [*Albinsson, 1991*] has been selected to be representative.

For lead, a K_d-value of 0.5 m^3/kg has been selected [*NAGRA, 1993*]. The same value applies for sorption in fresh and leached concrete, and in sand.

Americium, curium and europium

Americium and curium appear as trivalent ions and laboratory experiments demonstrate that americium and europium sorb strongly on cementious material. There is no literature data available on the sorption of curium, it should have the same sorption properties for curium as for americium and europium. A K_d-value of 5 m^3/kg has been selected to represent sorption of americium, curium and europium on fresh and leached concrete [*Allard et al., 1991, Nordén 1994*]. For degraded concrete and sand a value of 3 m^3/kg [*Allard et al., 1991, Albinsson, 1991*] has been selected to be representative.

Technetium, zirconium, thorium, protactinium, uranium, neptunium and plutonium

Technetium is tetravalent under reducing repository conditions and a K_d-value of 2 m^3/kg should be representative for sorption of technetium on fresh and leached concrete [*Allard et al., 1991*]. The same value has been chosen for technetium sorption in sand [*Albinsson, 1991*]. Thorium is not important as a component of the waste according to the latest inventory [*Lindgren et al., 1994*], but still it has been included in Table 7.1. The main reason for including thorium is that it has a stable tetravalent state and is insensitive to the relevant redox conditions and therefore a good "model" for all the tetravalent actinides.

The actinides are mainly tetravalent under repository conditions. However, due to the redox chemistry of plutonium and the low Eh, a fraction of dissolved plutonium is likely to be trivalent. This does not affect the sorption properties because a trivalent actinide has about the same strong sorption in cementitious material as the tetravalent actinides. Inorganic complexing agents in the groundwater or pore water are not expected, to influence the sorption of the tetravalent actinides. The K_d-value 5 m^3/kg has been selected to represent sorption of thorium, protactinium, uranium, neptunium, plutonium and zirconium on fresh and leached concrete [*Allard et al., 1991, Bradbury and Sarott, 1994*].

Data for sorption in granite at high pH [*Albinsson, 1991*] indicate that sorption in sand is strong and therefore a K_d-value of 5 m^3/kg has been chosen to be representative for the actinides and 2 m^3/kg for zirconium.

Niobium

Niobium exists as pentavalent $Nb(OH)_6^-$ over the whole pH range (fresh to degraded concrete) and is not expected to be dependent on the redox potential in the repository [*Baes and Mesmer, 1976*]. The strong hydrolysis should cause a similar sorption behaviour as for tetravalent technetium. The K_d-value 2 m^3/kg is taken from *Allard et al.* [*1991*].

Carbon

Carbon as carbonate will precipitate as $CaCO_3$ in concrete. In degraded concrete the calcium concentration has decreased substantially and the effect of carbonate precipitation is less significant. The K_d-values of 5, 1 and 0.1 m^3/kg have been selected to represent carbon sorption in fresh and leached concrete respectively [*Allard et al., 1991*]. For sorption in sand the K_d-value decreases to 0.01 m^3/kg [*Albinsson, 1991*]. Other forms of carbon, e.g. organic compounds like methane etc., have not been considered in this study.

Iodine

The sorption of iodine is significant in fresh concrete, but decreases in leached concrete. The K_d-values of 0.03, 0.003 and 0 m^3/kg has been selected to represent iodine sorption in fresh and leached and degraded concrete respectively [*Allard et al., 1991*]. For sorption in sand, 0 m^3/kg has been selected [*Albinsson, 1991*].

Tritium

Tritium will be present in the aqueous phase as dissolved gas or as tritiated water and will not exhibit true sorption. There is, however, a possibility that tritium will undergo isotope exchange reactions with bound water, hydroxide ions and protons. The effect is difficult to quantify and as a conservative choice, the sorption value is set to be zero for both concrete and sand.

Table 7.1 Selected representative distribution coefficients for concrete materials and sand.

		K_d[m^3/kg]			
		Concrete Materials			
Ox.State	**Element**	**Fresh concrete**	**Leached concrete**	**Degraded concrete**	**Sand**
	pH	13.5	12.5	10	8-10
M(I)	Cs[a,d]	0.002	0.1	0.1	0.05
M(II)	Sr[a,d]	0.002	0.02	0.02	0.01
	Ni[b,d], Co[c], Fe[c]	0.1	0.1	0.02	0.02
	Ra[b,d], Be[c], Cd[c]	0.05	0.05	0.05	0.1
	Pb[e]	0.5	0.5	0.5	0.5
M(III)	Am[a,d], Cm[a,d], Eu[f,d]	5	5	3	3
M(IV)	Np[b,d], Pa[b,c], Pu[a], Th[b,d], U[e,d]	5	5	5	5
	Tc[a,d]	2	2	2	2
	Zr[b,d]	5	5	5	2
M(V)	Nb[a]	2	2	2	2
Others	C (inorg.) [a]	5	1	0.1	0.001
	C (org.) [a]	0	0	0	0
	I[a]	0.03	0.003	0	0
	H[b,c]	0	0	0	0

a [*Allard et al., 1991*]
b [*Bradbury and Sarott, 1994*]
c authors' selection
d [*Albinsson, 1991*]
e [*NAGRA, 1993*]
f [*Nordén, 1994*]

7.1.2 Sorption in bentonite and sand/bentonite mixtures

Many of the above mentioned aspects of sorption of radionuclides in concrete also apply to sorption in a sand/bentonite mixture in a cementitious system. The pH of bentonite porewater is buffered by the bentonite minerals. However the surrounding concrete has enough capacity to release hydroxide ions so that in theory all the buffer capacity of bentonite is used. If that stage is reached, the pH of the clay will start to increase. The redox chemistry of the bentonite is not expected to be influenced. Some of the redox buffering capacity will be used by trapped air but after closure all air will be consumed for example by corrosion and a low Eh will be established in the clay. Therefore, the redox sensitive elements are expected to be in their lower oxidation state .

The use of sorption data from batch experiments to estimate the retention in bentonite and sand/bentonite mixtures is questionable. The degree of compactation influence the transport rate of radionuclides through bentonite. Whether this is solely due to different porosity or whether the sorption properties of the clay also are dependent on compactation is difficult to clarify [*Wanner et al., 1994*]. Also, sorption data from batch experiments are derived from a water/solid ratio which is much different from actual conditions. Therefore, values from diffusion experiments have to be used in order to calculate realistic distribution coefficients. Experimental diffusivity values [*Johnston et al., 1984, Albinsson et al., 1989, Kulmala and Hakanen, 1993*] have been used to select realistic apparent diffusivities. The diffusivities are listed in Table 7.2. The values presented for the diffusion in sand/bentonite mixture (90/10) are one order of magnitude higher than for pure bentonite. These values can be applied to a 85/15 mixture with negligible errors [*Albinsson and Engkvist, 1989, Johnston et al., 1984*]. Diffusion experiments indicate that the accessible porosity in the bentonite for anions is about 5% of the total porosity [*Brandberg and Skagius, 1991*]. For this study the effective diffusivities have been assumed to be 10^{-11} m^2/s for cations and 10^{-12} m^2/s for anions due to anion exclusion.

Table 7.2 Apparent diffusivities for radionuclides in bentonite and sand/bentonite (90/10). The distribution coefficients for bentonite and sand/bentonite mixtures can be calculated from the apparent and effective diffusivities [Johnston et al., 1984, Albinsson and Engkvist, 1989, Kulmala and Hakanen, 1993].

		Apparent diffusivity, $D_a(m^2/s)^{*)}$	
Ox.Stat	**Element**	**Pure bentonite**	**90% sand + 10% bentonite**
M(I)	Cs	10^{-12}	10^{-11}
M(II)	Sr, Ra	10^{-12}	10^{-11}
	Be, Cd, Co, Fe, Ni	10^{-13}	10^{-12}
M(III)	Am, Cm, Eu	10^{-14}	10^{-13}
M(IV)	Np, Pa, Pu, Tc, Th, U, Zr	10^{-15}	10^{-14}
M(V)	Nb	10^{-15}	10^{-14}
Other	I	10^{-11}	10^{-10}

*) Effective diffusivity, D_e, equal to 10^{-12} m^2/s for anions (I^-) and 10^{-11} m^2/s for cations.

The evaluation of the diffusive transport through the bentonite backfill barrier has highlighted a principal difficulty. It is customary to make a distinction between the two different cases: instationary diffusion ruled by the apparent diffusion constant D_a and stationary diffusion described by the effective diffusion constant D_e When estimating K_d-values from measured D_a values selection of conservative values of D_e can cause an overestimate of the K_d-values for sorption, leading to an overestimate of the barrier retardation. Therefor it is essential that the evaluation of K_d-values from experiments are made with consistent sets of diffusivities.

7.2 Solubility in groundwater and concrete porewater

In the near-field the solubility may, in addition to sorption, limit the element concentration in the pore water and thereby determine the source term release. The important aspects of water chemistry are pH, redox conditions and the presence of complexing agents. In this section the potential solubility of different elements in an alkaline/neutral and reducing water, which is representative of the waste environment in the repository, is discussed.

The redox conditions in the repository are expected to stabilise the tetravalent state of the actinides U, Pa, Np and Pu. Although the pH is high, no enhancement of the solubility of the tetravalent actinides is expected because of their inability to form negatively charged hydroxo complexes [*Engkvist, 1993*]. However, the di- and trivalent elements will form a significant fraction of these complexes and therefore hydrolysis of di- and trivalent actinide cations such as Am^{3+} and Cm^{3+} has to be considered when estimating their solubility values. The solubility for the actinides in Table 7.3 is based on a crystalline solubility limiting phase. For the other elements the solubility is based on an amorphous hydroxide limiting phase.

The concentration of CO_3^{2-} will be low due to the low solubility of $CaCO_3$ (pH 13.5, $[CO_3^{2-}] \sim 1.4 \cdot 10^{-4}$ M and pH 12.5, $[CO_3^{2-}] \sim 1.0 \cdot 10^{-4}$ M) and therefore the complexation of the metals with carbonate is expected to be negligible compared to complex formation with hydroxide which has a relatively high concentration due to the high pH.

7.2.1 Solubility of metal elements

Cesium

The quantities of cesium are small compared to the amount of water in the repository. Since the solubility of cesium is high, no solubility limit has been considered. The pH and Eh have no influence on the solubility of cesium in the repository.

Strontium

The quantities of strontium are small compared to the amount of water in the repository. Since the solubility is relatively high, no solubility limit has been considered.

Radium, cadmium, nickel, beryllium, iron, cobalt and lead

The solubility of radium is expected to be even higher than that of strontium due to the fact that the alkaline-earth metals exhibit less and less tendency to hydrolyse as the atomic number increases [*Baes and Mesmer, 1976*].

Cadmium is fairly soluble in the low pH range ($pH < 10$) but reaches a minimum solubility of less than $10^{-6.5}$ M at $pH \approx 11$. The solubility increases slowly above pH 12 and at pH 13 the solubility is 10^{-6} M [*Baes and Mesmer, 1976*].

Nickel has a low solubility in alkaline media with a minimum solubility of $10^{-7.5}$ M at pH 12-12.5. The solubility of nickel increases below pH 12 and above 12.5. However, for the relevant range of pH-values the solubility does not exceed 10^{-7} M [*Pilkington and Stone, 1990*].

Beryllium has a low solubility in neutral or slightly basic media. The minimum solubility is $10^{-6.7}$ M. The solubility is however, strongly enhanced by the formation of $Be(OH)_3^-$ and $Be(OH)_4^{2-}$ when pH exceeds 9 [*Baes and Mesmer, 1976*]. At pH 12.5 the solubility of Be reaches $10^{-3.7}$ M. No anions but the hydroxide ions present in the groundwater or the pore water of concrete and bentonite have an effect on the solubility of beryllium and cadmium [*Smith and Martell, 1976*].

Iron(II) has a minimum solubility of $< 10^{-6}$ M at pH 10.5 and the solubility increases to 10^{-5} M at pH 13 [*Baes and Mesmer, 1976*].

Cobalt has less tendency to form anionic hydroxide species and therefore the minium solubility is maintained in the pH region 10-12.5 at a value of $< 10^{-6}$ M [*Baes and Mesmer, 1976*].

The solubility of lead is strongly dependent on the pH conditions. At pH 10, the solubility is about 10^{-2} M but decreases to the minimum 10^{-3} M at pH 11. At higher pH the solubility slowly increases to 10^{-2} M at pH 13 [*Baes and Mesmer, 1976*].

Americium and curium

Americium and curium are strongly solubility limited in concrete influenced water in the repository. The solubility does not exceed 10^{-8} M in the pH range 8-13.5 (assuming no anionic complexes formed) [*Allard and Torstenfelt, 1985*].

Technetium, zirconium, thorium, protactinium, uranium, neptunium and plutonium

Measurements have demonstrated that tetravalent technetium has a low solubility in neutral to alkaline media [*Eriksen et al., 1992*]. The solubility in the repository is expected to be 10^{-7} M.

The solubility of zirconium is affected by the formation of anionic hydroxo species. The minimum solubility is less than 10^{-10} at pH 5 to 6, but increases slowly to $10^{-4.5}$ at pH 13. The solubility is based on ZrO_2 as limiting phase [*Baes and Mesmer, 1976*].

Thorium has a very limited solubility in neutral to alkaline solutions. The selected solubility is $10^{-9.5}$ M over the pH range 8 to 13.5 [*Baes and Mesmer, 1976*].

Tetravalent actinides do not form anionic complexes even at high alkalinity [*Engkvist, 1993*]. Since the carbonate concentration is low in cementious systems, the solubility for Pa, U, Np and Pu will not exceed 10^{-10} M [*Allard and Torstenfelt, 1985*].

Niobium

Niobium will dissolve only as pentavalent ions and the solubility will be limited to 10^{-9} M [*Kulmala and Hakanen, 1993*].

Table 7.3 Solubility limits in concrete porewaters and groundwater.

		Solubility in mol/l		
		Concrete porewater		**Groundwater**
Ox.State	**Element**	**pH 13.5-12.5**	**pH 10**	**pH 8-10**
M(II)	Be[a]	10^{-3}	10^{-6}	10^{-6}
	Fe[a]	10^{-5}	10^{-6}	10^{-3}
	Co[a]	10^{-5}	10^{-6}	10^{-3}
	Ni[b]	10^{-7}	10^{-7}	10^{-5}
	Cd[a,c]	10^{-6}	10^{-6}	10^{-2}
	Pb[a]	10^{-2}	10^{-2}	$>10^{0}$
M(III)	Am[c]	10^{-8}	10^{-8}	10^{-8}
	Cm[c]*	10^{-8}	10^{-8}	10^{-8}
M(IV)	Tc[d]	10^{-7}	10^{-7}	10^{-7}
	Zr[a,c]	$10^{-4.5}$	$10^{-6.5}$	10^{-9}
	Th[a,c]	$10^{-9.5}$	$10^{-9.5}$	$10^{-9.5}$
	Pa[a]	10^{-10}	10^{-10}	10^{-10}
	U[c]	10^{-10}	10^{-10}	10^{-10}
	Np[c]	10^{-10}	10^{-10}	10^{-10}
	Pu[c]	10^{-10}	10^{-10}	10^{-10}
M(V)	Nb[d]	10^{-9}	10^{-9}	10^{-9}

* In analogy with Am.
a [*Baes and Mesmer, 1976*]
b [*Pilkington and Stone, 1990*]
c [*Allard and Torstenfelt, 1985*]
d [*Kulmala and Hakanen, 1993*]

7.3 Coprecipitation and adsorption on corrosion products

In SFL 3-5 large amounts of metal waste will be stored. The metals mainly consist of different qualities of steel. With time, the metals will corrode and produce different solid corrosion products. The corrosion products expected to be formed in largest quantities are different oxides/hydroxides of iron. Such compounds are well known to adsorb and coprecipitate other metal ions present in solution. It has therefore been decided to briefly study these phenomena for the SFL 3-5 repository. A more detailed description on coprecipitation processes is given in Appendix B.

In steel waste neutron activated isotopes of nickel may form coprecipitates with ferric hydroxides formed by corrosion of steel. Due to the resemblance in chemical properties between iron and nickel, it is likely that recrystallisation of ferric hydroxide may be accompanied by similar recrystallisation of nickel. If this is true, increasingly stable solid solutions may be obtained. This might significantly reduce the release of nickel, especially from SFL 5.

The potential importance of coprecipitation can be illustrated by application of Eq. 6 in Appendix B to estimate the reduction of the release of nickel. The steel waste primarily consists of stainless steel with slightly less than 70% iron and about 10% nickel. The steel waste container also has a similar composition. The molar ratio of nickel to iron in a hypothetical coprecipitate with ferric iron, resulting from congruent corrosion of the steel, is on the order of 0.14:1. From Eq. 6 in Appendix B the solubility of nickel would then be expected to be reduced to about 0.14 times the solubility of a corresponding pure nickel precipitate.

In an experimental investigation the coprecipitation of nickel with iron has been studied [*Thornber and Wildman, 1984*]. Under the studied conditions it was found that the nickel solubility is lower than for pure nickel phases. It was also indicated that coprecipitates with nickel were stable at pH 8 and higher, with nickel solubility decreasing with time during the experiments.

The radionuclide release calculations have shown that Ni-59, which is expected to be solubility limited, may be the dose dominant radionuclide after approximately 10^5 years. Consequently, a reduction of the solubility of nickel by coprecipitation with ferric iron could directly influence the radionuclide release. Coprecipitation has, however, not been considered in the release calculations in the prestudy.

In the SFL 3-5 repository also other coprecipitation processes may be important. Among these the precipitation of calcite/aragonite and brucite are likely to occur when deep groundwater is mixed with the alkaline water in the repository (as a result of equilibration with concrete in construction materials). Also the recrystallisation of different cement minerals may capture certain radionuclides, but little is known on this subject.

7.4 Content of inactive elements and potential influence

The waste to be stored in SFL 3-5 will comprise both inactive and active species of the same element. The content of inactive elements in the waste may be of importance for the evaluation of the source term releases. Especially for metal waste with induced activity, where many of the radionuclides in the waste originates from irradiation of the elements in the metals in addition to possible surface contamination.

Therefore estimates have been performed on ratios between active and inactive nickel and zirconium respectively by looking at some SFL 5 waste types with high induced activity; core grid, SIRM-guide tubes and the fuel boxes [*Lindgren and Pers, 1994*].

The core grids are made of stainless steel AISI 304, with a nickel content of 10 % by weight. The reported induced activity of Ni-59 is about 1.4 g/kg and of Ni-63 is 0.33 g/kg. The content of Ni-59 is estimated to be about 1.4% and the content of Ni-63 about 0.33% of the nickel content in the core grids.

The SIRM-guide tubes are made of stainless steel SIS 2353, with a nickel content of 13% by weight. The reported induced activity, Ni-59 about 2.9 g/kg and Ni-63 about

0.81 g/kg, are higher than in the core grids. The content of Ni-59 is estimated to be about 2.2% and the content of Ni-63 about 0.6% of the nickel content in the SIRM-guide.

The examples given are nickel containing metal parts with a high rate of induced activity. Most of the metal waste in SFL 5 will have less induced activity and lower ratio active to inactive nickel. The total activity content of Ni-59 and Ni-63 in the SFL 5 waste will be determined by the induced activity. The surface contamination of Ni-59 and Ni-63 will only contribute marginally to the total activity content in the SFL 5 waste.

The BWR fuel-boxes are made of Zircaloy 4, with a zirconium content of 98.5% by weight. The reported induced activity of Zr-93 is 0.58 g/kg, i.e. 0.06% of the initial content of zirconium is activated to Zr-93. The total activity content of Zr-93 in the SFL 3-5 waste is determined by the induced activity in the fuel boxes allocated to SFL 5. The contribution of Zr-93 from surface contamination is several orders of magnitude lower than the induced activity.

To summarize we conclude that only part of the element content of nickel and zirconium in the studied metal parts are transformed to radioactive isotopes of nickel and zirconium by neutron irradiation, even though these parts have high induced activities. Based on the estimated ratios, we think it feasible to assume a ratio between active and inactive nickel of 1% in stainless steel and zirconium of 0.1% in Zircaloy. This is of importance for the release of radioactive isotopes of nickel and zirconium if these elements become solubility limited in the repository or if the release is determined by the corrosion mechanism. For metal parts with less induced activity the effect will be pronounced.

7.5 Influence of complexing agents and organic material on sorption and solubilities

The various categories of waste in SFL will contain considerable amounts of organic material, such as ion exchange resins, plastic and rubber etc, as well as cellulose (see Chapter 3). Essentially all of the organic components, totally some 400 tonnes, will be placed in SFL 3 (Table 3.5).

The degradation of the organic components under the conditions in the repository (anaerobic; pH above 12) will generate a fraction of soluble organic compounds that may have a significant influence on the solubility and adsorption behaviour of the radionuclides and thereby their releases and migration out of the repository. The expected decomposition of ion-exchange resins, as well as various natural and synthetic polymers, plasticizers, hydrocarbons etc has previously been discussed in several reports [*Allard and Borén, 1991, and references quoted therein*]. A general conclusion, also stated in a recent review [*Atkinson et al., 1993*], is that the degradation products from these organics would have only a minor influence on the releases of radionuclides from the repository, with the exception of certain categories of extremely strong complexing agents like the aminopolycarboxylic acids (NTA, EDTA, DTPA) [*Allard and Persson, 1985*]. However, these complexing agents are not expected in anything but very minor quantities in the waste.

The alkaline degradation of cellulose, however, will yield products that may have large effects on solubility as well as adsorption of certain elements. The first reported observations on the enhanced solubility of plutonium in the presence of cellulose at high pH were made in 1986 [*Bradshaw et al., 1986, Bradshaw et al., 1987*] and these observations have been at least qualitatively confirmed by several groups in the UK, Switzerland and Sweden.

Inorganic complexing agents are in comparison less important, but with one exception and that is cyanides. This has been observed by [*Bradbury and Sarott, 1994*] and in the Swiss safety assessment of a LLW and ILW repository in Wellenberg, equal weight was given to cyanides and cellulose [*NAGRA, 1994*]. Cyanides are present in some of the concrete conditioned waste from Studsvik, where Cs has been coprecipitated with ferrocyanides (copper(II)-iron-cyanide) from waste solutions [*Lindgren et al., 1994*]. This waste will go to SFL 3. However, the estimated quantities of cyanides are only 10% of the amount of cellulose in SFR 3 [*Lindgren et al., 1994*]. Given equal weight as complexing agents it should be enough for the prestudy to restrict the discussion to cellulose and therefore only effects of cellulose degradation will be discussed in the subsequent sections.

7.5.1 Alkaline degradation of cellulose

Various assumptions have been made concerning the composition of the cellulose degradation products that would account for the observed enhanced solubility of e.g. plutonium. It is known that minor fractions of polyhydroxycarboxylic acids (typically hexose derivatives) can be formed. Several hexose derivatives have been selected as model substances in experimental studies of cellulose degradation effects on radionuclide mobility [*Greenfield et al., 1993a*].

Recent experiments have confirmed, that D-glucoisosaccharinic acid is the dominating strong complexing acid that will be formed during alkaline cellulose degradation [*Greenfield et al., 1993b*], just as was previously suggested [*Allard and Borén, 1991, Van Loon, 1993*]. Concentrations of isosaccharinic acid of up to the mM-range are feasible. The further decomposition of isosaccharinic acid is not known, and steady-state concentration levels under continuous generation from cellulose and subsequent degradation can not be assessed. Total concentrations of up to 0.01 M of acidic degradation products, corresponding to a total organic carbon concentration in solution of some 600 mg/l appears to be possible at high initial cellulose loading.

7.5.2 Effects of cellulose on radionuclide solubility and adsorption

There are numerous studies of particularly plutonium solubility in the presence of cellulose or hexose derivatives, but also of Th, U, Np, Am, Tc, Ni [*Greenfield et al. 1993b*]. There a few studies on adsorption of particularly Pu, but also Am, Th, Ra and Eu, on cement and on minerals at various pH, in the presence of cellulose degradation products or hexose derivatives (D-gluconic acid, D-glucoisosaccharinic acid) [*Bradbury and Sarott, 1994*]. There are also several efforts to develop models that can be used for the calculation of the effects of cellulose on the adsorption, primarily based on solubility studies in the presence of gluconic acid and isosaccharinic acid, with efforts to verify the models with data from the few adsorption studies performed.

It can be concluded, considering recent results on the adsorption of Pu, Th and Eu at pH 12 on cement and mineral surfaces [*Baston et al., 1994, Nordén and Allard, 1994*] that:

- The presence of cellulose degradation will enhance the solubility of particularly tetravalent elements at high pH (as well as the presence of strong complexing acids among the hexose derivatives).

- The major potential complexing acid that can be formed at substantial yield by the alkaline degradation of cellulose is D-glucoisosaccharinic acid.

- The presence of cellulose degradation products at levels of 20-30 mg/l or above may lead to an initial reduction of the adsorption of tri- and tetravalent elements on mineral surfaces at high pH (e.g. cement at pH 12).

- The presence of the acidic hexose derivatives D-gluconic acid at concentrations above 10^{-4} M and D-glucoisosaccharinic acid (at slightly higher concentrations) would lead to an initial reduction of the adsorption, just as for a real mixture of cellulose degradation products.

- The reduction of adsorption due to the formation of complexes with hexose derivatives etc can not be calculated from the corresponding increased solubility; evidently the complexes are to a significant extent sorbed on the solid surface as well.

- Initial effects on adsorption due to gluconic or isosaccharinic acids is decreasing with time at pH above 12, particularly for isosaccharinic acid. The effects are only minor after 5-6 months (gluconic acid) and 1-2 months (isosaccharinic acid) at initial concentrations in the mM-range.

7.5.3 Application to the SFL 3-5 repository

All cellulose will be found in SFL 3:

Cellulose	46 tonnes (+ other organics)
Steel	210 tonnes (+ other metals)
Concrete/cement	9860 tonnes (waste packages)
Water	5300 tonnes

Thus, an average cellulose/cement ratio of 0.5% and a cellulose/water ratio of 0.9% can be assumed. These ratios are far below most reported conditions in laboratory studies but in reasonable agreement with studies in progress [*Nordén and Allard, 1994*]. A maximum TOC-concentration of around 700 mg/l would be expected, with a corresponding content of up to $5 \cdot 10^{-3}$ M isosaccharinic acid (including other strong acids, if any) based on comparison of adsorption data for systems with cellulose and with pure acids. An initial reduction of the adsorption of a trivalent element by a factor of 200 (in terms of the distribution coefficient) is feasible in a worst case scenario (data for D-gluconic acid), which is reduced to about 50 after 2 weeks and to negligible effects after about 6 months [*Allard et al, 1995*].

It is reasonable to neglect the effects of cellulose degradation on **radionuclide adsorption** as the central case in a performance assessment of SFL, considering the low loading (in terms of cellulose/cement as well as cellulose/water ratios) and assuming that the hold-up time in the high pH-region (above 12) would be at least years. The fairly conservative choice of distribution coefficients from the critical tri- and tetravalent elements (Table 7.1) would probably include the uncertainties due to minor amounts of complexing organics. However, the effects of organics on **solubilities** can not *a priori* be neglected. This is a reason to disregard solubility constraints for the tetravalent radionuclides (Th, U, Np and Pu) in a case where cellulose dehydration products are expected to be present.

Questions that still have to be addressed are, however:

- What are the yields of strong complexing agents under the present conditions - and how fast is the subsequent degradation of the D-glucoisosaccharinic acids?
- What are the long-term effects of the high concentrations of calcium for the reduction of any effects from complexing organics (as well as possibly of iron from the corrosion).

7.6 Colloid generation and transport

Colloidal matter, typically in the size range 100-400 nm, is present at low levels in essentially all natural waters. Undisturbed groundwaters are likely to contain particulate colloidal matter (silicates, possibly metal hydroxides and calcium carbonate) at levels up to 0.1 mg/l [*Ledin, 1993*]. Changes in redox conditions as well as in temperature or carbon dioxide pressure may lead to the formation of colloidal Fe-hydroxides and calcium carbonate. Coagulation of high molecular weight organics as well as certain colloidal silicates can occur in a salinity gradient. An underground repository can in principle serve as a source of colloids, e.g. from the degrading concrete and corroding metal components as well as from backfill materials.

7.6.1 Radionuclide adsorption on colloidal matter

The adsorption of radionuclides on colloidal matter would be similar to the adsorption on macroscopic surfaces of the same mineral [*Ramsey et al., 1991, Ledin, 1993*]. Thus, the radionuclides that are released from the waste forms will be distributed among all available surfaces, governed by the hydrochemical conditions as well as the composition and physical properties of the solid phases. Thus, the presence of a significant colloidal phase could enhance the apparent concentration of radionuclides in the fluid phase above what could be expected considering solubility constraints and adsorption/distribution processes.

7.6.2 Radionuclide transport by colloids

The backfill (bentonite) as well as cementitious materials (waste forms and containers) are the most likely sources of colloids in the repository. The concentrations of colloids from these sources can presently not be assessed. However, it can be assumed that:

- both cement and clay colloidal matter would carry a negative surface charge, just as all the major solid surfaces (e.g. concrete) in the repository.
- larger particles than 100-200 nm would probably not pass through the intact cement or the backfill zone which would act as mechanical "filters".

The formation of a substantial colloidal fraction in the size range below 100 nm must be assumed until experimental evidences indicate other conditions. The effects of high calcium concentrations would have a destabilizing effect which can not presently be quantified. It must be assumed that a significant fraction of particularly the tri- and tetravalent radionuclides that are released from the waste form will be associated with colloidal matter **within** the waste containers. However, it is unlikely that negatively charged particulate matter of sizes below 100 nm would easily enter into a pore system (in the concrete or backfill) where all solid surfaces also carry negative charges (c.f. the low diffusivity observed for the anionic I^- in clay and rock systems [*Torstenfelt, 1986*].

7.6.3 Application to the SFL 3-5 repository

The most probable case for SFL 3-5 is that colloids are formed and exist at considerable concentration within containers of concrete or containing cementitious waste forms. It is also probable that the backfill materials can serve as a potential colloid source. It is likely that only a minor fraction could be transported through concrete and/or backfill. The best estimate must presently be that the quantitative transport of radionuclides out of the near-field can be neglected. Experiments should be performed to clarify this matter and to verify this hypothesis.

8 Hydraulic and thermal conditions in the repository

The estimated groundwater flow in the different repository parts SFL 3, 4 and 5, and the increase in temperature due to radioactive decay of radionuclides in the waste are presented in this chapter. The calculations are further described in [*Lindgren and Pers, 1994*].

8.1 Groundwater flow in SFL 3-5

The water flow inside the vaults has been estimated with an analytical model. The model is made for calculations of the direction and size of the water flow through a body with contrasting hydraulic conductivity compared to the surrounding medium. The two media is assumed to be homogeneous and the vault is presented by a long prolate spheroid (cigar).

The water flux in undisturbed rock is assumed to be 0.1 l/m^2, year, see Section 5.1. The flow inside the vault is directly proportional to the flux in the rock.

Groundwater flow in SFL 3

SFL 3 is planned to be backfilled with bentonite and sand/bentonite. As long as the bentonite and sand/bentonite is intact, which has been assumed in this study, the hydraulic conductivity will be low, equal or less than in the rock. This means that it can be assumed that the release with water flow is negligible in comparison to release with diffusion. Therefore no efforts has been made in this study to calculate the water flow in SFL 3.

Groundwater flow in SFL 4

Different alternatives of the extension of the SFL 4 vault exists. The vault has in the calculations been regarded as two vaults, each with a length of 280 m and a cross section area of 35 m^2.

The water flow in the vault was calculated as a function of the average hydraulic conductivity in the vault. The calculations were performed for different combinations of hydraulic gradient and average hydraulic conductivity in the rock. An example of the water flow as a function of the average hydraulic conductivity in the vault is given in Figure 8.1. Two cases are depicted; water flow parallel to the vault and water flow perpendicular to the vault, these cases are extreme so all other results will fall in between. The figure shows that the average hydraulic conductivity must be less than about 10^{-5} m/s to limit the flow inside the vault. Since SFL 4 will be backfilled with sand, the average hydraulic conductivity in the vault will be so low that it may limit the flow. However, the maximum flow is chosen in order not to underestimate the flow. The total flow in the two SFL 4 vaults is then 3.4 m^3/year.

Groundwater flow in SFL 5

SFL 5 consists of three equal vaults. The length of each vault is 130 m and the cross section area is 70 m^2.

The calculations were made in the same way as for SFL 4. The result indicate that the flow inside the vault approaches the value for completely empty vault even for rather low average hydraulic conductivities inside the vault. Therefore the maximum level of the flow was chosen. The total flow in all the three SFL 5 vaults together is 1.8 m^3/year.

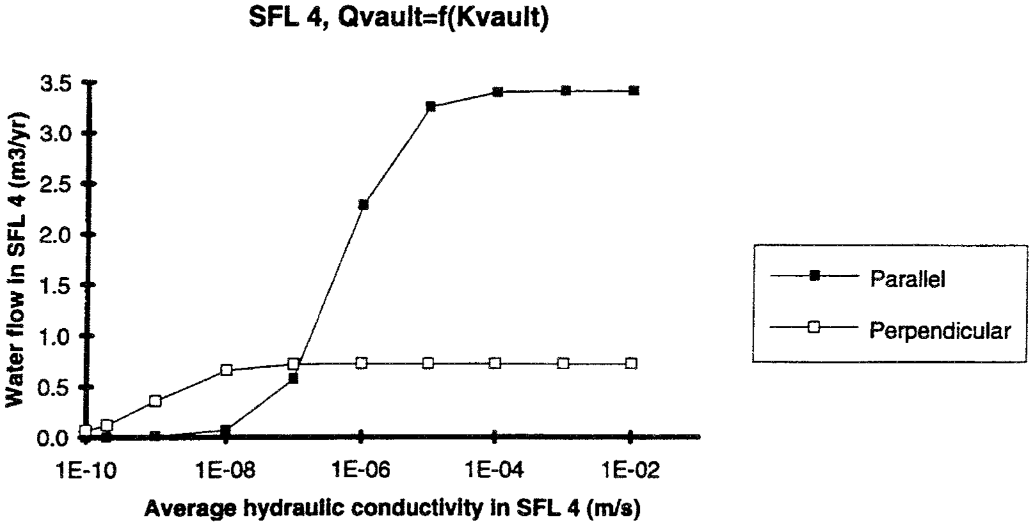

Figure 8.1 *Water flow in SFL 4 as a function of the average hydraulic conductivity in the vault (Hydraulic conductivity in the rock = 10^{-9} m/s, hydraulic gradient in the rock = 0.003, corresponding to a water flux of 0.1 l/m^2, year).*

8.2 Estimates of influence of waste on temperature

Heat evolution in a repository may cause thermally induced water flow and influence the long-term properties of the barriers, among other things. Therefore it is important to examine the potential temperature increase in a repository. One source of heat evolution is heat being generated from the waste by the loss of energy when the radionuclides decays. The increase in temperature due to radionuclide decay has been calculated for the repository with the highest radionuclide content, i.e. SFL 5.

In a set of simplified calculations, the geometry of the influenced area has been described by a cylinder. The inner cylinder, with a radius of 3 m, represents the waste. The first shell, 0.5 m thick, represents the concrete (package wall and tunnel walls). The outer shell, 12 m thick, represents the rock outside the tunnels. These 12 m represent half the distance between two tunnels, i.e. at this distance a no flow boundary can be prescribed. The calculations show that the total increase in average temperature after 100 years is 1.8 °C. Co-60 and Ni-63 is totally dominating, 99% of the increase in temperature arise from decay of these two nuclides.

A more detailed calculation including the two dominating nuclides Co-60 and Ni-63 was also performed in order to calculate the maximum temperature in the waste. This calculation shows that the heat transport is fast in comparison to the heat generation so the temperature inside the waste is negligible higher than the average temperature.

The conclusion from these calculations is that the increase in temperature due to radionuclide decay is only some few degrees in SFL 5 and can therefore be neglected. The radionuclide content in the two other repository parts, SFL 3 and SFL 4, is much lower than in SFL 5 and it is therefore concluded that the heat generation from the waste is negligible also for these repository parts.

9 Gas formation

Gas may be formed in the repository. The main sources are corrosion of metals and microbial degradation of organic materials. Large volumes of gas can at least potentially be created by these processes. Thermal degradation and chemical reactions of materials as well as radiolytic decomposition in radioactive waste repositories can possibly also generate gaseous compounds which contributes to the gas formation. A proper understanding of gas formation processes, gas transport mechanisms and effects of gas formation is important for the evaluation of the performance of a repository.

9.1 Gas formation sources and mechanisms

The main types of gas formation processes are:

- Corrosion of steel and other metals
- Microbial degradation of organic materials
- Radiolytic decomposition

Corrosion of metals

Steel is the major metallic material present in the repository, but there are also other metals present, such as aluminium and Zircaloy. In a short term perspective the corrosion of aluminium may not be neglected, since significant amounts are present. However, in a long-term perspective the principal source of gas from metals is anaerobic corrosion of steel, yielding hydrogen gas.

The chemical environment in the repository is determined by the presence of large amounts of cement, for instance in concrete structures, backfill materials, and waste conditioning materials. Besides dissolved oxygen, other oxidising agents present in the pore water, or in the waste and cement materials, may change the redox conditions and thereby influence the corrosion mechanisms. In addition, the presence of certain species in the pore water, primarily chloride, can change the stability of passivating corrosion products, and thereby alter the initiation of the corrosion processes. The presence of some alloying metals in steel, e.g. nickel and chromium, can increase the passivation of steel and hence decrease the corrosion rate. However, in reducing environments, the passivating layers may be less stable than in oxidising environments. For steel having a high content of chromium, the passivation may extend into the field of strongly reducing conditions.

Several other environmental conditions have potential impact on the corrosion rates and mechanisms, such as hydrogen pressure, carbonates, and microbes. Hydrogen evolving corrosion reactions are influenced by the hydrogen pressure. To inhibit the corrosion of steel would require an unacceptable high pressure build-up of hydrogen, but also low hydrogen pressures can have a passivating effect by decreasing the corrosion rate. In neutral and acidic solutions, high carbonate concentrations can increase the steel corrosion rate. In a cementitious environment, the carbonate concentration is determined by the solubility of calcite, and the effect on the corrosion rate is therefore expected to be minor.

Different species of microorganisms, such as bacteria and yeasts, may induce or enhance the corrosion of metals. However, the extent of corrosion caused by microbes may be limited due to the dependence on available supply of carbon sources, oxidants, nutrients etc.

Corrosion of steel

Hydrogen evolving corrosion can occur only in the absence of dissolved oxygen. That implies that anaerobic corrosion will start when the initially present oxygen has been consumed by aerobic corrosion or some other oxygen consuming reaction, such as microbial activity.

Under *aerobic conditions* steel corrodes in accordance with the following generalised reaction for different corrosion products:

$$2\ Fe(s) + 3/2\ O_2 + X\ H_2O \Rightarrow Fe_2O_3(s) \text{ or } 2Fe(OH)_3(s) \text{ or } 2\ FeO(OH)(s)$$

When *anaerobic conditions* prevail, the amount of hydrogen evolved can be estimated by the overall reaction for corrosion of steel, for which the formation of magnetite is the end product.

$$3\ Fe(s) + 4\ H_2O \Rightarrow Fe_3O_4 + 4\ H_2(g)$$

The corrosion rate for steel has been compiled from literature for different environmental conditions [*Höglund and Bengtsson, 1991*]. Corrosion rates of steel in anaerobic environments are usually within in the range of 10^{-7} to 10^{-5} m per year. This range was used for gas formation calculations in the safety assessment of SFR. In the prestudy of SFL 3-5 a corrosion rate for steel of 10^{-6} m/year has been assumed. This is the same value as in the Swiss SMA-study [*NAGRA, 1993*] and our selection of steel corrosion rate is based on the same references.

Corrosion of Zircaloy

Zircaloy is an alloy with approximately 98% of zirconium. Corrosion of zirconium can occur according to the following reaction:

$$Zr(s) + 2\ H_2O \Leftrightarrow 2\ H_2(g) + ZrO_2$$

In the prestudy a corrosion rate for Zircaloy of 10^{-8} m/year has been assumed. In the KBS-3 study a value of 10^{-7} m/year was used, but a later assessment of the corrosion of Zircaloy waste in concrete, by the Swiss organisation NAGRA, uses a value of 10^{-8} m/year.

Corrosion of aluminium

Aluminium is not thermodynamically stable in water, but has a very dense protective oxide layer. However, in alkaline environments this oxide layer will dissolve and a rapid corrosion of aluminium yielding hydrogen can take place according to the general reaction.

$$2\ Al(s) + 2\ OH^- + 4\ H_2O \Rightarrow 2\ AlO(OH)_2^- + 3\ H_2(g)$$

In alkaline environments aluminium corrodes rapidly and the corrosion rate can be in the range of 10^{-3} to 10^{-2} m per year. A corrosion rate of 10^{-3} m/year was has been assumed in prestudy of SFL 3-5, this value was also used for aluminium in the Swiss SMA-study [*NAGRA, 1993*].

Microbial degradation of organic material

The presence of different microorganisms in the repository can not be excluded. The organic content in some waste types may be digested by microorganisms which can generate gases. The gas is generated as a result of microbial oxidation of different organic components in the waste. The organic material is utilised as a carbon source by the microorganisms, whereas, different inorganic compounds may be used as oxidants: molecular oxygen, nitrate, sulphate or carbonate. In addition, microbial activity requires access to water and certain nutrients at trace levels.

The chemical environment significantly influences the rate of microbial degradation, However, the remarkable ability of many microorganisms to adapt to different environments must be respected. The surface area of the organic material available for microbial attack is important, bitumen and plastics may have a low surface to volume ratio, whereas cellulosic materials like paper have a very large surface area. Different types of organic substrates also give different degradation rates. Carbohydrates are relatively easily degraded, e.g. different sugars, whereas complex and halogenated aromatic hydrocarbons often are unaffected by microbial degradation.

Radiolytic decomposition

Radiolytic decomposition of materials in the repository with radioactive wastes may lead to generation of gases. During irradiation of materials by alpha, beta, and gamma radiation excited and ionised species are created. The excited species can in turn undergo a variety of chemical reactions. The gross effect of irradiation is proportional to the absorbed energy and depends also on the material composition.

Irradiation of water can lead to hydrogen generation. In pure water the final products are hydrogen and hydrogen peroxide. If multivalent metals ions are present in the water, many researchers have noted a decrease in the hydrogen production. The content of water in the irradiated materials significantly influences the radiolytic decomposition rates, since the generated free electrons in the water react with the molecules in the materials. During irradiation of concrete, hydrogen gas is formed mainly by decomposing the water present in the cement, whereas the major constituents of cement are stable. The hydrogen generation rate increases slightly with increasing water/cement ratios. Gas can also be produced by irradiation of organic hydrocarbons. The hydrogen gas is formed by breaking the bond between the carbon and hydrogen atoms. Small amounts of carbon monoxide, carbon dioxide and methane may also be created. The formation of carbon oxides will consume already present oxygen or oxygen created by radiolytic decomposition. Radiolytic decomposition may not only lead to the formation of gases but also to compounds influencing the speciation of radionuclides.

The effect of irradiation is commonly expressed as the radiolytic yield, the G-value. The G-value is defined as the number of molecules produced per 100 eV of absorbed energy by the material. The G-value for hydrogen generation in pure water is about 0.5 molecules per 100 eV for beta- and gamma-irradiation and about 1.5 molecules per 100 eV for alpha irradiation [*Höglund and Bengtsson, 1991*]. In the prestudy radiolytic yields for pure water has conservatively been assumed.

9.2 Gas formation in SFL 3-5

Gas formation can occur by several mechanisms as described in the previous section. Gas formation due to radiolysis of water, microbial degradation of organics and corrosion of metals has been estimated in this prestudy [*Lindgren and Pers, 1994*]. Only the amounts of gas are calculated and no consequences of the produced gas are included.

Gas formation in SFL 3

The gas formation due to radiolysis of water was calculated for SFL 5 and was found to be small. Since the radionuclide content is much less in SFL 3 it can be concluded that the amount of gas produced will be of minor importance and no calculations were performed.

Organics are present in SFL 3. The estimated amount of cellulose is 45 920 kg and of other organic materials 340 000 kg, of which the dominating part is ion-exchange resins. The gas formation rate due to microbial degradation of the organic materials was calculated using gas formation rates of 0.7 moles/(kg.year). for cellulose degradation and 0.05 moles/(kg.year) for degradation of other organics, and assuming that 50% of the gas found is insoluable [*Wiborgh et al., 1986*].This results in an initial gas formation rate of 551 m^3(STP)/year, which corresponds to 11 m^3/year at repositing depth. The contribution from cellulose degradation is about 7 m^3/year and from degradation of other organics about 4 m^3/year. The maximum yields for cellulose degradation and degradation of ion-exchange resins are reported in [*Brandberg and Wiborgh, 1982*] to be 37 moles/kg for cellulose and 30 moles/kg for ion-exchange resins. Using these values, gas formation due to degradation of cellulose will prevail for 53 years and degradation of other organics will generate gas during 600 years. Thus, the initial gas formation rate of 11 m^3 /year will after 53 years decrease to about 4 m^3/year and terminate after 600 years

The gas formation due to corrosion of steel was calculated with a corrosion rate of 10^{-6} m/year. The total surface area of steel in the waste is 9948 m^2, which gives a gas production rate of 42 m^3(STP)/year. According to this rate the corrosion of the steel waste will take about 3000 years. The steel in the waste packaging (steel containers, concrete reinforcement etc.) has a total surface area of 48 619 m^2, resulting in a gas production rate of 204 m^3(STP)/year. The time for total corrosion of the waste packaging will be about 2000 years.

The gas formation due to corrosion of aluminium was calculated with a corrosion rate of 10^{-3} m/year. The total surface area of aluminium is 5802 m^2, resulting in a gas production rate of 19 500 m^3(STP)/year. The calculated time for total corrosion of aluminium is about 3 years. It is possible that some corrosion will occur before repository closure and that the gas production will be lower. It is also likely that the corrosion will not start or proceed uniformly and as a result of that the peak of generation is expected to be spread out over more than 3 years.

Gas formation in SFL 4

The gas formation due to radiolysis of water was calculated for SFL 5 and was found to be small. Since the radionuclide content is much less in SFL 4 it can be concluded that the amount of gas produced will be of minor importance and no calculations were performed.

The gas formation due to corrosion of steel was calculated with a corrosion rate of 10^{-6} m/year. The total surface area of steel in the waste is $2.4 \cdot 10^5$ m^2, which gives a gas production rate of 1008 m^3(STP)/year. According to this rate the corrosion of the steel waste will take about 5000 years. The steel in the waste packaging has a total surface area of 44 200 m^2, resulting in a gas production rate of 186 m^3(STP)/year. The time for total corrosion of the waste packaging will be about 3000 years.

Gas formation in SFL 5

The gas formation due to radiolysis of water is directly correlated to the radionuclide inventory. The gas formation rate will be largest in the beginning when most decay occur. The average rate during the first 10 years after repository closure is 0.004 m^3(STP)/year and package, i.e. a total gas production of 55 m^3(STP) during the first 10 years in SFL 5. The gas production during these first 10 years is totally dominated by decay of Co-60 and Ni-63. The total amount of gas produced by radiolysis will be 964 m^3(STP).

The gas formation due to corrosion of steel was calculated with a corrosion rate of 10^{-6} m/year. The average surface area of the steel waste in a package is 26 m^2, resulting in a gas production rate of 0.11 m^3(STP)/year. The time for corrosion of the steel in the waste is about 7000 years. The surface areas of the steel in the packaging in SFL 5 are 13.9 m^2 reinforcement/package and 25.5 m^2 cassette/package, resulting in a gas production rate of 0.17 m^3(STP)/year and package. The time for corrosion of the reinforcement is about 4000 years and for the cassettes about 3000 years. The total gas production rate for corrosion of steel in SFL 5 is 377 m^3(STP)/year.

The gas formation due to corrosion of Zircaloy was calculated with a corrosion rate of 10^{-8} m/year. Zircaloy is only present in some waste types and the total surface area is 4005 m^2. The total gas production rate in SFL 5 due to corrosion of Zircaloy is 0.13 m^3(STP)/year and the total time for corrosion will be about 120 000 years.

Summary of gas formation calculations

In the prestudy only the potential amounts of gas formed from waste and waste packaging in SFL 3-5 have been estimated. In later assessments the contribution from repository structures to the total gas formation rates and the potential influence of gas on contaminated release from the repository must be evaluated.

A summary of the estimated gas formation rates, due to radiolysis, microbial degradation of organic material in the waste, corrosion of steel waste and packagings, and corrosion of aluminium and Zircaloy in the waste, is given in Table 9.1.

Table 9.1 Summary of the gas formation rates in SFL 3, SFL 4 and SFL 5.

	Gas formation rate (m^3/year)[1]		
	SFL 3	**SFL 4**	**SFL 5**
Radiolysis	-	-	0.1[2]
Microbial degradation		-	-
cellulose	7		
other organics	4		
Corrosion of steel			
waste	1	20	3
packaging	4	4	5
Corrosion of Al	390[3]	-	-
Corrosion of Zircaloy		-	0.003
Total	*406*	*24*	*8*

1) Corresponding volumes at repository conditions (10°C, 50 atm).
2) Average rate during the first 10 years, the rate decreases with time.
3) Depleted after about 3 years

10 Estimates of near-field releases

10.1 General

Two modelling approaches were used to calculate the release from the near-field of SFL 3-5. To get a first indication of the release levels and of important radionuclides a model was used which neglects all transport resistances in the waste packages, concrete structures and sand/gravel backfill, a so called "stirred tank" model. Calculations were performed for all radionuclides in the present inventory for which chemical and physical data are available.

Release calculations for the defined Reference Case of the Reference Scenario, see Chapter 6, were performed with a more detailed model considering advective and diffusive transport through the barriers in the repository. Release calculations were performed for a subset of radionuclides which were judged to be representative with respect to their abundance, mobility and dose contribution. This model was also used to estimate the release of lead and beryllium from SFL 3 to exemplify the release of chemotoxic elements from the repository.

To get a measure of the radiotoxicity of the near-field release, the calculated radionuclide releases from the near-field were converted to intermediate doses by assuming that the total release from the repository is captured in a well with a capacity of 2000 m^3/year and that an individual consumes 0.6 m^3 of this well water annually. The dose conversion factors for ingestion used are from [*NRPB-R245, 1991*] (Table 3.8).

The near-field release calculations and conversion to intermediate doses are summarised in the following sections. A more detailed description is found in [*Lindgren and Pers, 1994*].

10.2 Calculations with the stirred tank model

10.2.1 Modelling approach

The basic assumptions in the "stirred tank" model is that the radionuclides are immediately dissolved and evenly distributed inside the repository vault. The resulting concentration in the water is determined by the available water volume and sorption on the barrier materials. If this concentration exceeds the solubility of the radionuclide then the concentration is assumed to be equal to the solubility. The distribution coefficients, K_d, and the solubilities are given in Tables 7.1 and 7.3. The release rates from the vaults are calculated as the product of the concentration and the water flow through the repository vault.

In SFL 3, diffusive transport dominates over advective transport through the bentonite barriers. The diffusive transport was expressed as an equivalent water flow which accounts for the diffusion resistance in the bentonite barriers and the mass transfer resistance between the bentonite barriers and the water flowing in the surrounding rock. The radionuclide concentrations in the water inside the bentonite barriers were calculated by assuming that all radionuclides were immediately dissolved in the water inside the bentonite barriers and sorbed onto all concrete in the vault, i.e. waste matrix, packages and backfill, concrete structures and porous concrete backfill. To account for pH decrease due to leaching of the concrete, but still not underestimate the release, the lowest values of the distribution coefficients of fresh and leached concrete were chosen (see Table 7.1). The calculated concentrations were compared with solubility values corresponding to concrete porewater (see Table 7.3). For none of the radionuclides the calculated concentration exceeded the highest of these solubilities.

The water flow releasing the radionuclides from SFL 4 was assumed to be equal to the calculated maximum water flow through the tunnel system, 3.4 m^3/year (see Chapter 8). In the present design of the SFL 4, the amount of concrete is small. Therefore the radionuclide concentrations were calculated by assuming that all radionuclides were immediately dissolved in the water in the vault and sorbed onto the sand/gravel backfill in the vault. To consider potential effects on pH in the vault of the concrete present the lowest values of the distribution coefficients of sand and concrete were chosen (see Table 7.1). The calculated concentration did not exceed the solubility limits in the pH-range 10-13.5 for any of the radionuclides.

In SFL 5, the water flow releasing the radionuclides was assumed to be equal to the calculated maximum water flow through the vaults, 1.8 m^3/year (0.6 m^3/year through each vault, see Chapter 8). The radionuclide concentrations in the vault were calculated assuming immediate dissolution of all radionuclides in the water in the vaults and sorption onto all concrete in the vaults, and onto the sand/gravel backfill. The calculated concentrations were compared with solubilities in concrete porewaters (see Table 7.3). The fact that only a small fraction of nickel and zirconium in the waste is active isotopes, 1% and 0.1% respectively, was also considered. The calculated concentrations did not exceed the solubility for any of the radionuclides except for nickel. The concentration of radioactive nickel isotopes was then set to 1% of the solubility.

10.2.2 Results

The maximum release rates calculated with the stirred tank model are given in Table 10.1. Since the transport resistances in the barrier materials are neglected in the model, no time for radioactive decay in the barrier is allowed and the maximum release rates are obtained immediately. This means that the stirred tank model will give fairly good estimates of the release rates of long-lived radionuclides, but largely over-predicts the release rates of short-lived radionuclides. Since most of the radionuclides with the highest calculated release rates have half-lifes of less than 100 years, the time delay in the barriers will play an important role in limiting the release of these radionuclides.

Table 10.1 Maximum release rates, stirred tank model.

Radionuclide	**Half-life (year)**	**Maximum release rate (Bq/year)**		
		SFL 3	**SFL 4**	**SFL 5**
H-3	$1.2 \cdot 10^{1}$	$5 \cdot 10^{10}$	$5 \cdot 10^{5}$	$4 \cdot 10^{11}$
Be-10	$1.6 \cdot 10^{6}$			$1 \cdot 10^{5}$
C-14[1)]	$5.7 \cdot 10^{3}$	$3 \cdot 10^{4}$		$4 \cdot 10^{6}$
Fe-55	2.7	$2 \cdot 10^{4}$	$8 \cdot 10^{8}$	$2 \cdot 10^{8}$
Ni-59	$7.5 \cdot 10^{4}$	$3 \cdot 10^{5}$	$3 \cdot 10^{5}$	$3 \cdot 10^{5*}$
Co-60	5.3	$3 \cdot 10^{6}$	$5 \cdot 10^{8}$	$2 \cdot 10^{9}$
Ni-63	$9.6 \cdot 10^{1}$	$2 \cdot 10^{7}$	$5 \cdot 10^{7}$	$3 \cdot 10^{7*}$
Sr-90	$2.9 \cdot 10^{1}$	$1 \cdot 10^{6}$	$8 \cdot 10^{7}$	$7 \cdot 10^{6}$
Zr-93	$1.5 \cdot 10^{6}$	<1	2	$2 \cdot 10^{4}$
Nb-93m	$1.4 \cdot 10^{1}$	<1	$2 \cdot 10^{1}$	$4 \cdot 10^{4}$
Nb-94	$2.0 \cdot 10^{4}$	$1 \cdot 10^{1}$	$5 \cdot 10^{1}$	$6 \cdot 10^{4}$
Tc-99	$2.1 \cdot 10^{5}$	5	5	$7 \cdot 10^{3}$
I-129	$1.6 \cdot 10^{7}$	$2 \cdot 10^{1}$	$1 \cdot 10^{2}$	4
Cs-134	2.1	$5 \cdot 10^{3}$	$5 \cdot 10^{7}$	$4 \cdot 10^{2}$
Cs-135	$2.3 \cdot 10^{6}$	$3 \cdot 10^{2}$	$1 \cdot 10^{3}$	$2 \cdot 10^{2}$
Cs-137	$3.0 \cdot 10^{1}$	$3 \cdot 10^{7}$	$8 \cdot 10^{7}$	$7 \cdot 10^{6}$
Eu-152	$1.3 \cdot 10^{1}$	$2 \cdot 10^{1}$		
Eu-154	8.8	3		
Eu-155	5.0	3		
Pb-210	$2.2 \cdot 10^{1}$	$5 \cdot 10^{3}$		
Ra-226	$1.6 \cdot 10^{3}$	$6 \cdot 10^{4}$		
Ac-227	$2.2 \cdot 10^{1}$	$1 \cdot 10^{2}$		
Pa-231	$3.3 \cdot 10^{4}$	<1		
Th-230	$7.7 \cdot 10^{4}$	<1		<1
Th-232	$1.4 \cdot 10^{10}$	2		
U-234	$2.4 \cdot 10^{5}$	<1		<1
U-235	$7.0 \cdot 10^{8}$	5		
U-236	$2.3 \cdot 10^{7}$	<1		<1
U-238	$4.5 \cdot 10^{9}$	$1 \cdot 10^{1}$		
Np-237	$2.1 \cdot 10^{6}$	<1		<1
Pu-238	$8.8 \cdot 10^{1}$	$8 \cdot 10^{1}$	6	<1
Pu-239	$2.4 \cdot 10^{4}$	$1 \cdot 10^{3}$	1	<1
Pu-240	$6.5 \cdot 10^{3}$	$3 \cdot 10^{2}$	1	<1
Pu-241	$1.4 \cdot 10^{1}$	$1 \cdot 10^{3}$	$4 \cdot 10^{2}$	$2 \cdot 10^{1}$
Am-241	$4.3 \cdot 10^{2}$	$1 \cdot 10^{3}$	<1	2
Am-243	$7.4 \cdot 10^{3}$	<1	<1	<1
Cm-243	$2.9 \cdot 10^{1}$	<1	4	<1
Cm-244	$1.8 \cdot 10^{1}$	6	1	<1

[1)] *total inventory assumed to be inorganic C-14*

* *release is limited by solubility*

10.3 Reference Case calculations

10.3.1 Modelling approach

To get a better estimate of the near-field release rates, a more detailed model of the barrier systems in the three repository parts were used in the Reference Case calculations. The calculations were carried out by the computer code TRUMP [*Edwards, 1969*] considering advective and diffusive transport through the barriers in the repository. Radioactive decay, but no chain decay was considered. The calculations were performed for a subset of radionuclides which were judged to be of importance based on the results from the calculations with the "stirred tank" model as well as to be representative what concerns retention and radiotoxicity. The selected nuclides and their initial activity in the waste are given in Table 10.2.

The basic premises for the Reference Case calculations are that the repository has become fully resaturated and that reducing conditions have been re-established. Furthermore, both the hydraulic situation and groundwater chemistry in surrounding rock are assumed to be stable over the time period of interest.

Table 10.2 *Initial activity (Bq) in waste of radionuclides selected for the Reference Case calculations.*

Radionuclide	**Half-life (year)**	**Initial activity in waste (Bq)**		
		SFL 3	**SFL 4**	**SFL 5**
Ni-59	$7.5\cdot10^{4}$	$9.4\cdot10^{12}$	$3.8\cdot10^{10}$	$1.2\cdot10^{15}$
Co-60	5.3	$8.3\cdot10^{13}$	$5.4\cdot10^{13}$	$5.2\cdot10^{15}$
Ni-63	$9.6\cdot10^{1}$	$8.0\cdot10^{14}$	$6.0\cdot10^{12}$	$1.1\cdot10^{17}$
Zr-93	$1.5\cdot10^{6}$	$4.7\cdot10^{7}$	$5.4\cdot10^{7}$	$2.2\cdot10^{12}$
Nb-93m	$1.4\cdot10^{1}$	$1.9\cdot10^{7}$	$2.6\cdot10^{8}$	$1.9\cdot10^{12}$
Nb-94	$2.0\cdot10^{4}$	$9.2\cdot10^{9}$	$5.5\cdot10^{8}$	$3.1\cdot10^{12}$
I-129	$1.6\cdot10^{7}$	$1.9\cdot10^{7}$	$4.4\cdot10^{5}$	$3.1\cdot10^{5}$
Cs-134	2.1	$3.6\cdot10^{9}$	$7.1\cdot10^{11}$	$2.1\cdot10^{7}$
Cs-135	$2.3\cdot10^{6}$	$1.9\cdot10^{8}$	$1.6\cdot10^{7}$	$1.1\cdot10^{7}$
Cs-137	$3.0\cdot10^{1}$	$2.1\cdot10^{13}$	$1.1\cdot10^{12}$	$3.9\cdot10^{11}$
Pu-238	$8.8\cdot10^{1}$	$1.3\cdot10^{11}$	$1.8\cdot10^{8}$	$9.8\cdot10^{7}$
Pu-239	$2.4\cdot10^{4}$	$2.1\cdot10^{12}$	$3.4\cdot10^{7}$	$2.4\cdot10^{7}$
Pu-240	$6.5\cdot10^{3}$	$4.6\cdot10^{11}$	$3.9\cdot10^{7}$	$2.7\cdot10^{7}$
Pu-241	$1.4\cdot10^{1}$	$2.2\cdot10^{12}$	$1.2\cdot10^{10}$	$1.9\cdot10^{9}$
Am-241	$4.3\cdot10^{2}$	$1.6\cdot10^{12}$	$1.2\cdot10^{7}$	$2.1\cdot10^{8}$

SFL 3

The waste allocated to SFL 3 is a mixture of various materials, e.g. activated and surface contaminated scrap metals, precipitates, ashes, ion-exchange resins, glove boxes, radiation sources, laboratory outfit and radiation protection equipments. Steel drums or concrete containers are the dominating waste packaging types (see Chapter 3).

The waste packages are assumed to be filled with water initially, and a high pH has been established due to the presence of cement and concrete. Radionuclides in the waste are assumed to be immediately dissolved and evenly distributed inside the waste packages, i.e. in the waste, conditioning material and packaging material. The initial concentration in the pore water in the waste packages is determined by the total radionuclide content in the waste, the pore water volume and by sorption onto cement and concrete in the waste packages. Due to the slow leaching of the cement and concrete (see Chapter 4) the lowest values of the distribution coefficients representative for fresh and leached concrete given in Table 7.1 were chosen. The calculated concentrations were compared with solubilities representative for concrete environments (see Table 7.3). For none of the radionuclides the calculated concentration exceeded the highest value of these solubilities.

The radionuclide migration from the waste packages is assumed to be dominated by diffusion through the porous concrete backfill surrounding the packages, the concrete compartment walls and the bentonite and sand/bentonite barriers. Sorption in the barriers as well as diffusion into and sorption on internal concrete structures will delay the release of radionuclides from the vault.

The model used to calculate the migration from the waste packages is schematically depicted in Figure 10.1. The model is a semi two-dimensional description of the barrier system where the diffusion through the top, bottom and sides of the vault are described as three parallel, one-dimensional pathways. The total release is the sum of the contributions from these pathways. The waste packages provide no resistance to transport of dissolved radionuclides. Due to the geometry of the vault the area of the barriers accessible to diffusion will increase with distance from the packages. In the model this is described by a stepwise area increase between each barrier, where the area of each barrier is estimated from the thickness and volume of the barrier. Diffusion into porous concrete between the waste packages and the internal structures and into the internal structures as well as diffusion into porous concrete between the packages are described as dead-end pathways in the model. The transfer of radionuclides from the bentonite and sand/bentonite barriers is expressed in the model as an equivalent water flow, Q_{eq}, which accounts for the diffusion resistance between the clay barriers and the slowly moving groundwater in fractures in the surrounding rock.

Effective diffusivities, porosities and densities used in the calculations are given in Table 4.4. Due to the slow leaching of the concrete (see Chapter 4) the lowest values of the distribution coefficients representative for fresh and leached concrete given in Table 7.1 were chosen. The distribution coefficients for bentonite and sand/bentonite were calculated from the apparent and effective diffusivities given in Table 7.2.

The equivalent water flow, Q_{eq}, is dependent on the magnitude and direction, perpendicular or parallel to the axis of the vault, of the water flow in the near-field rock. Assuming the same water flow as in the undisturbed far-field rock the equivalent water flow was calculated to be 0.05 m^3/year for a flow direction parallel to the vault axis and 0.08 m^3/year for a flow direction perpendicular to the vault axis. Assuming that excavation effects will increase the flow in the near-field rock by a factor of 10 resulted in an equivalent water flow of 0.2 and 0.3 m^3/year for a flow direction parallel and perpendicular to the vault axis, respectively. Because of uncertainties in the effects of excavation an equivalent water flow of 0.1 m^3/year was selected in the calculations of the radionuclide release.

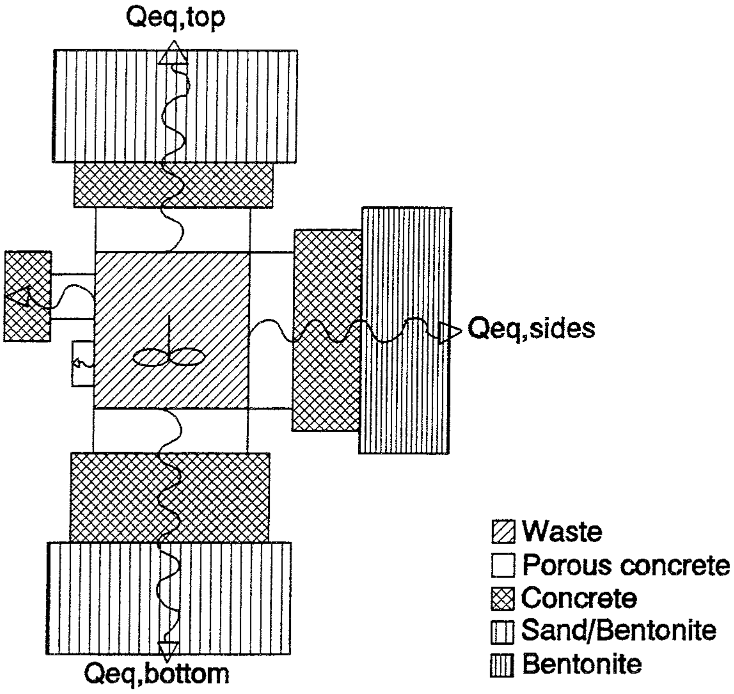

Figure 10.1 *Schematic illustration of the SFL 3 release model (not to scale). Shown are the diffusion pathways through the top, bottom and sides of the repository vault. Diffusion into porous concrete between waste packages as well as into porous concrete and internal structures are shown as dead-ends to the left in the figure.*

SFL 4

The waste to be disposed of in SFL 4 is decommissioning waste, transport casks and containers, and fuel storage canisters (see Chapter 3). The main waste materials are metals and concrete. The potential packagings for the waste are steel vessels and concrete vessels. In the calculations it is assumed that steel vessels will be used.

The steel vessels are assumed to be filled with water initially, and radionuclides in the waste are immediately dissolved in the water in the packages. Due to corrosion of the steel vessels and the metal waste it is assumed that a low redox potential is maintained in the packages and the presence of concrete waste may create an alkaline environment. The radionuclide concentration is assumed to be determined by the radionuclide content and the water volume in the vessels. The calculated concentrations were compared with the solubilities given in Table 7.3 as representative for concrete porewaters and saline groundwater. It was found that the calculated initial concentration was lower than the solubility for all radionuclides studied. Potential effects of sorption on concrete waste materials are disregarded because of the small amount of concrete waste present. Corrosion limited release of radionuclides present as induced activity in metal waste is also disregarded since the induced activity content is low compared to the surface bound activity.

The steel vessels are assumed to be permeable enough to allow water flowing in the surrounding sand/gravel backfill to also flow through the vessels. Dissolved radionuclides leaving the vessels with the flowing water are further transported with the moving water and by diffusion through the sand/gravel backfill into the near-field rock. Sorption on the sand/gravel backfill will delay the release of the radionuclides.

The SFL 4 release model is schematically depicted in Figure 10.2. The waste packages provide no resistance to transport of dissolved radionuclides and is modelled as a "stirred tank". The release model is one-dimensional. Therefore the length of the pathway through the surrounding sand/gravel backfill is set to the average of the pathway length in different directions, and the area of the backfill through which transport occurs is calculated from the total backfill volume and the average pathway length.

Effective diffusivities, porosities and densities used in the calculations are given in Table 4.4. The sorption capacity of the backfill is dependent on the composition of the water in the vault. If the water composition is affected by the presence of concrete, the distribution coefficients in the backfill are similar to those for concrete (see Chapter 7). Although it does not seems very likely that the conditions in the backfill will be similar to a fresh or leached concrete environment, the lowest of the distribution coefficients given in Table 7.1 for conditions ranging from groundwater to fresh concrete porewater were selected in the calculations. Of the radionuclides studied, only the cesium isotopes have lower distribution coefficients in a fresh and leached concrete environment than in a groundwater system. Consequently, the selected distribution coefficient for the cesium isotopes in the backfill may well be too pessimistic and the resulting release to the near-field rock over estimated. The water flow through the backfill was assumed to be equal to the calculated maximum water flow through the tunnel system, 3.4 m^3/year (see Chapter 8).

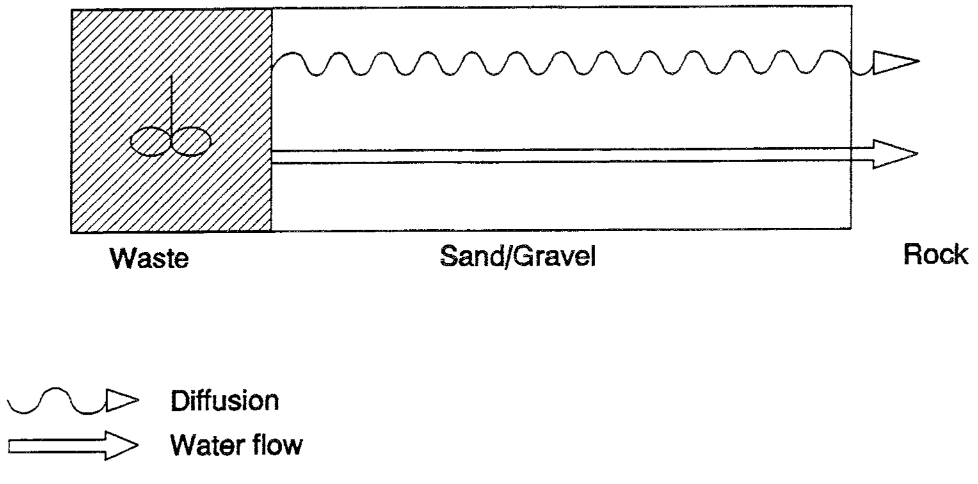

Figure 10.2 Schematic illustration of the SFL 4 release model (not to scale).

SFL 5

SFL 5 is the repository part intended for reactor internals, i.e. metal components with both induced activity and surface contamination. Reinforced concrete containers with inner steel cadettes will be used as waste packagings (see Chapter 3).

The waste packages are assumed to be filled with water initially. Due to corrosion of the steel cadettes and metal waste it is assumed that a low redox potential is maintained in the packages and that a high pH has been established due to the presence of cement and concrete. Radionuclides are assumed to be immediately dissolved and evenly distributed in the water in the waste and concrete backfill in the waste packages. The initial concentration in the water is determined by the total radionuclide content in the waste, the water volume and by sorption onto the concrete backfill in the waste packages. Due to the slow leaching of the cement and concrete (see Chapter 4) the lowest values of the distribution coefficients representative for fresh and leached concrete given in Table 7.1 were chosen. The calculated concentrations were compared with solubility values representative for a fresh and a leached concrete environment (see Table 7.3). The fact that only small fractions of nickel and zirconium in the waste are radioactive isotopes, 1% and 0.1% respectively, was also considered. The calculated concentrations did not exceed the solubility for any of the radionuclides except for nickel and zirconium. The concentration of radioactive nickel and zirconium isotopes was then set to the solubility lowered by the isotopic dilution factor.

The concentration in the water in the waste packages of activation products in the metal waste may be limited by the corrosion of the metals. To investigate this calculations were performed where it was assumed that steel corrosion determines the release to the water in the waste packages of the radionuclides Ni-59, Ni-63 and Nb-94 and that Zircaloy corrosion determines the release of Zr-93 and Nb-93m. The corrosion rates used were 10^{-6} m/year for steel and 10^{-8} m/year for Zircaloy.

Dissolved radionuclides in the water in the waste packages may be transported by diffusion and with moving water through the concrete packaging and surrounding compartment structures, and captured by water flowing in the sand/gravel backfill surrounding the compartments. Further transport through the backfill to the near-field rock is assumed to occur with moving water and by diffusion. Sorption in the concrete packaging, concrete structures and in the sand/gravel backfill will delay the release of radionuclides to the near-field rock.

A calculation was made to assess whether diffusive or advective transport dominates the transport through the concrete packages and the concrete structures. A comparison of the flow resistances in the sand/gravel backfill and the concrete barriers showed that the flow resistance in the concrete barriers are several orders of magnitude higher than in the sand/gravel backfill. A negligible part of the water entering the vault will then flow through the concrete barriers. It was also demonstrated that diffusion is the dominating transport mechanism in the concrete barriers even if 1% of the water entering the vault flows through the concrete barriers.

The SFL 5 release model used in the calculations are schematically depicted in Figure 10.3. Dissolved radionuclides in the backfill in the waste packages are transported by diffusion through the concrete container walls and the concrete structures and by diffusion and moving water in the surrounding sand/gravel backfill. Due to the geometry of the vault the area of the barriers through which transport occurs will increase with distance from the packages. In the model this is described by a stepwise area increase between each barrier, where the area of each barrier are estimated from the thickness and volume of the barrier. Diffusion into concrete container walls which are facing other concrete containers is included in the model as dead-end paths.

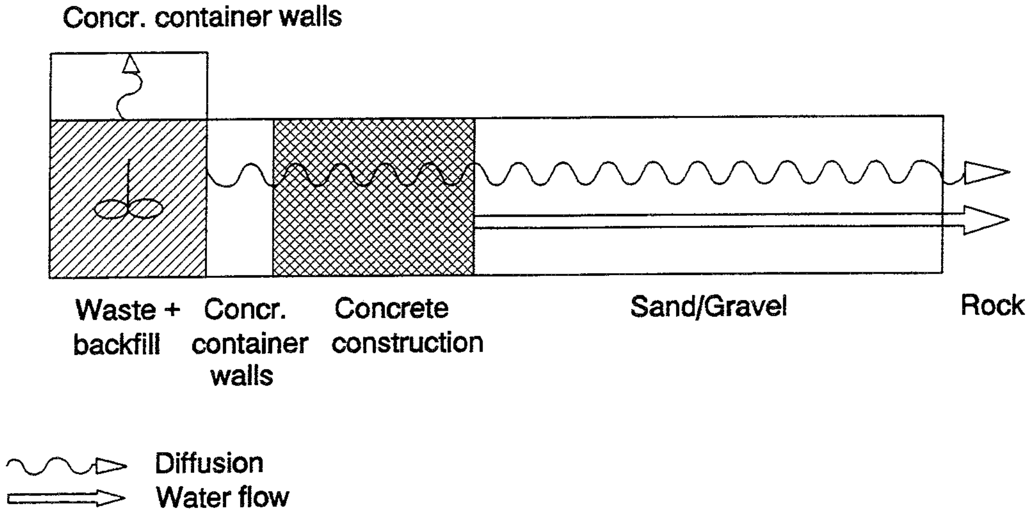

Figure 10.3 Schematic illustration of the SFL 5 release model (not to scale and the increase in surface area is not shown).

Effective diffusivities, porosities and densities used in the calculations are given in Table 4.4. Due to the slow leaching of the concrete (see Chapter 4) the lowest values of the distribution coefficients representative for fresh and leached concrete given in Table 7.1 were chosen. The same distribution coefficients were selected for the sand/gravel backfill since leaching of the concrete barriers will affect the conditions in the surrounding backfill. The water flow through the backfill was assumed to be equal to the calculated maximum water flow through the vaults, 1.8 m^3/year (0.6 m^3/year through each vault, see Chapter 8).

10.3.2 Radionuclide release from SFL 3

The radionuclide release rates from SFL 3 as a function of time are shown in Figure 10.4. Table 10.3 gives the maximum release rates and time for maximum release rates. Results are given only for those of the radionuclides considered in the Reference Case calculations that obtained a maximum release rate of 1 Bq/year and above. The total maximum release rate occurs after about 100 000 years and is dominated by Ni-59. The release rates of more short-lived radionuclides are much lower because of the retention and decay in the barrier system, even for low-sorbing elements like Cs. The maximum release rate of Cs-137 is about three orders of magnitude lower than for Cs-135 and more than five orders of magnitude lower than for Ni-59.

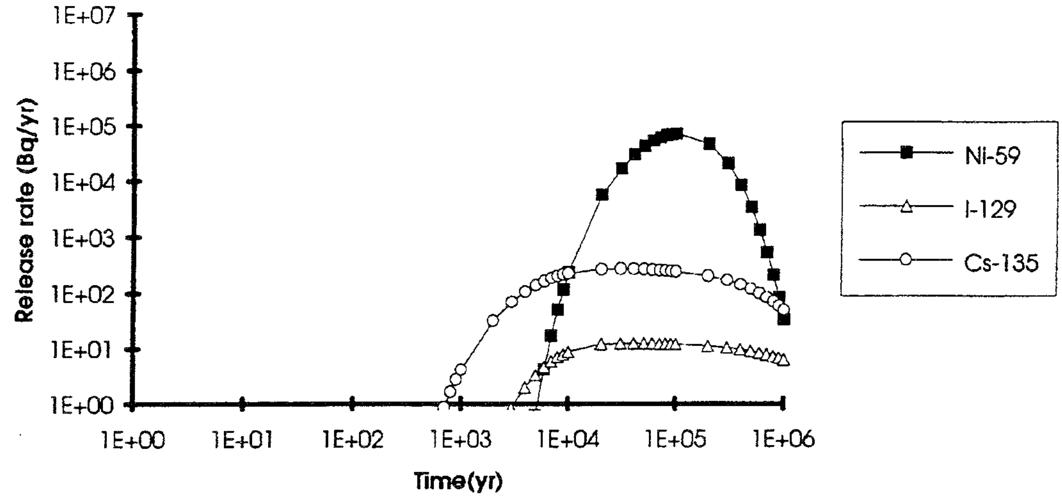

Figure 10.4 Radionuclide release from SFL 3, Reference Case.

Table 10.3 Maximum release rates of 1 Bq/year and above from SFL 3, Reference Case.

Radionuclide	**Half-life (year)**	**Time for max. release rate (year)**	**Maximum release rate (Bq/year)**
Ni-59	$7.5 \cdot 10^4$	$8 \cdot 10^4$	$7 \cdot 10^4$
I-129	$1.6 \cdot 10^7$	$3 \cdot 10^4$	$1 \cdot 10^1$
Cs-135	$2.3 \cdot 10^6$	$3 \cdot 10^4$	$3 \cdot 10^2$

As mentioned earlier, the calculations were performed for a subset of radionuclides in the waste inventory. However, the calculations with the simple "stirred tank" model were carried out for all radionuclides in the inventory for which data were available, and the results can be used to discuss the potential importance of radionuclides not included in the Reference Case calculations.

In the "stirred tank" model all transport resistances in the waste packages, concrete structures and sand/gravel backfill are neglected. This means that the model largely over-predicts the release of short-lived radionuclides since no time for decay is allowed in the barriers, while fairly good estimates of the maximum release rates for long-lived radionuclides are obtained. By considering the retention in the barriers the maximum release rates of short-lived nuclides like Co-60, Cs-137 and Ni-63 are decreased by several orders of magnitude to values below 1 Bq/year (see Tables 10.1 and 10.3). For Cs-135 and I-129, which are long-lived radionuclides with small retention in the barriers, the two models give almost the same maximum release rates. For Zr-93, which is a long-lived radionuclide with high retention in the barriers, the Reference Case maximum release rate is not obtained within 1 million years which is the maximum simulation time. However, the Reference Case maximum release rate of Zr-93 would not exceed the maximum release rate calculated by the "stirred tank" model which is below 1 Bq/year.

Some long-lived radionuclides which are not included in the Reference Case calculations are Tc-99, Th-230, Th-232, U-234, U-235, U-236, U-238 and Np-237. All these nuclides have higher retention in the barriers than Ni-59 and would then obtain their maximum release rates at a later time than Ni-59. The maximum release rate of these radionuclides would not exceed the maximum release rates calculated with the "stirred tank" model. For most of these radionuclides the expected maximum release rate would then be less than 1 Bq/year and none of the radionuclides would be released at a rate higher than 10 Bq/year (see Table 10.1). These values are well below the maximum release rates of Ni-59 and Cs-135 calculated with the Reference Case model (Table 10.3).

The results from the calculations with the "stirred tank" model shows that Ra-226 reaches a release rate level that is less than ten times lower than the release rate of Ni-59 (Table 10.1). The retention of Ra-226 in the barriers will be smaller than for Ni-59, but Ra-226 has a shorter half-life. Ra-226 will also be formed in the barriers due to chain decay. The magnitude of the maximum release rate of Ra-226 will probably be similar to the maximum release rates given in Table 10.3, but not likely exceed the maximum release rate calculated by the "stirred tank" model which is $6 \cdot 10^4$ Bq/year (Table 10.1).

Another radionuclide that may obtain a maximum release rate comparable to those given in Table 10.3 is C-14, provided that parts of it exists as organic carbon in the waste. Since organic carbon does not sorb in the barriers and C-14 has a half-life which is of the same magnitude as the time for maximum release of a slightly sorbing nuclide as Cs-135 (see Figure 10.4) the retention in the barrier system will not allow much decay of organic C-14. Another important factor which determines the maximum release level is, of course, the initial activity of organic carbon in the waste.

10.3.3 Radionuclide release from SFL 4

The radionuclide release rates from SFL 4 as a function of time are shown in Figure 10.5. Table 10.4 gives the maximum release rates and time for maximum release rates. Results are given only for those of the radionuclides considered in the Reference Case calculations that obtained a maximum release rate of 1 Bq/year and above. During the first 100 years, Cs-137 dominates the release. At longer times the two nickel isotopes, Ni-63 and Ni-59, are the dominating radionuclides. However, as mentioned previously in section 10.3.1, the choice of distribution coefficient for cesium in the sand/gravel backfill may be too pessimistic. A more appropriate choice of distribution coefficient would delay the release and decrease the maximum release rate of Cs-137.

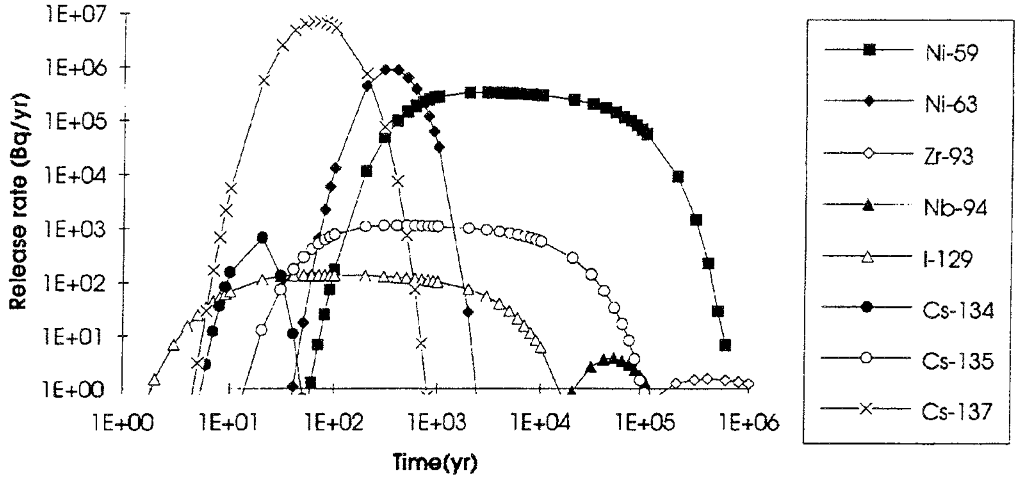

Figure 10.5 Radionuclide release from SFL 4, Reference Case.

Table 10.4 Maximum release rates of 1 Bq/year and above from SFL 4, Reference Case.

Radionuclide	Half-life (year)	Time for max release rate (year)	Maximum release rate (Bq/year)
Ni-59	$7.5 \cdot 10^4$	$2 \cdot 10^3$	$3 \cdot 10^5$
Co-60	5.3	$7 \cdot 10^1$	1
Ni-63	$9.6 \cdot 10^1$	$3 \cdot 10^2$	$9 \cdot 10^5$
Zr-93	$1.5 \cdot 10^6$	$4 \cdot 10^5$	2
Nb-94	$2.0 \cdot 10^4$	$5 \cdot 10^4$	4
I-129	$1.6 \cdot 10^7$	$6 \cdot 10^1$	$1 \cdot 10^2$
Cs-134	2.1	$2 \cdot 10^1$	$7 \cdot 10^2$
Cs-135	$2.3 \cdot 10^6$	$4 \cdot 10^2$	$1 \cdot 10^3$
Cs-137	$3.0 \cdot 10^1$	$7 \cdot 10^1$	$7 \cdot 10^6$

The radionuclide Sr-90 has not been included in the Reference Case calculations. However, in the calculations with the "stirred tank" model the calculated release rate of Sr-90 is the same as for Cs-137 (see Table 10.1). This result together with the similarity between Cs-137 and Sr-90 what regards half-life and sorption ability indicate that the maximum release rate of Sr-90 is approximately the same as for Cs-137.

Fe-55 is another short-lived radionuclide which has not been considered in the Reference Case calculations. This radionuclide has the same retention in the sand/gravel backfill and somewhat higher initial activity in the waste than Co-60, but shorter half-life. It is therefore expected that Fe-55 would obtain a maximum release rate which is lower than 1 Bq/year.

Based on the results obtained with the "stirred tank" model the release rates and doses for the more long-lived radionuclides not included in the Reference Case calculations are expected to be well below those shown in Figure 10.5.

10.3.4 Radionuclide release from SFL 5

The radionuclide release rates from SFL 5 as a function of time are shown in Figure 10.6. Table 10.5 gives the maximum release rates and time for maximum release rates. Results are given only for those of the radionuclides considered in the Reference Case calculations that obtained a maximum release rate of 1 Bq/year and above. As in SFL 4, Cs-137 will dominate the release rate at early times, but at a lower maximum level than in SFL 4. In the time interval 1000 to 10 000 years after repository closure the dominating contribution is from Cs-135 and in the later part also from Ni-59 which continues to dominate at times longer than 10 000 years.

The effect of solubility limitations and isotopic dilution of the nickel isotopes is that the maximum release rates are lowered by three orders of magnitude. The release of both Nb-94 and Zr-93 is limited by the corrosion rate of the metals. The effect on the release of Nb-94 is small. The release is 10% lower if corrosion of the steel determines the dissolution rate compared to when all Nb-94 in the steel waste is immediately dissolved. A larger effect was found for Zr-93, where the slow corrosion of Zircaloy lowers the release by almost two orders of magnitude. The release rate of Ni-59 given in Table 10.5 is the solubility limited release and the values given for Zr-93 and Nb-94 are the maximum release rates obtained when corrosion limits the release.

Sr-90 is one of the radionuclides not considered in the Reference Case calculations that is expected to give a major contribution to the release at earlier times. The reasons for that are the same as in SFL 4, the same calculated release with the "stirred tank" model (see Table 10.1) and the similarity between Sr-90 and Cs-137 what concerns retention in the barriers and half-life.

Another radionuclide not considered in the Reference Case calculations which may contribute to the release during early times is H-3. The reason for that is the expected small retention in the barrier system for non-sorbing nuclides like H-3. This is illustrated by the release curve for the slightly sorbing nuclide Cs-137 which is released at a rate of 1 Bq/year already after about 20 years and obtain its maximum release after about 100 years (Figure 10.6).

The results from the calculations with the "stirred tank" model indicate that none of the more long-lived and sorbing nuclides not considered in the Reference Case calculations would reach release rate levels that are comparable to those given in Table 10.5. One possible exception is Tc-99. However, the maximum release rate will not exceed the value obtained in the calculations with the "stirred tank" model which is $7 \cdot 10^3$ Bq/year.

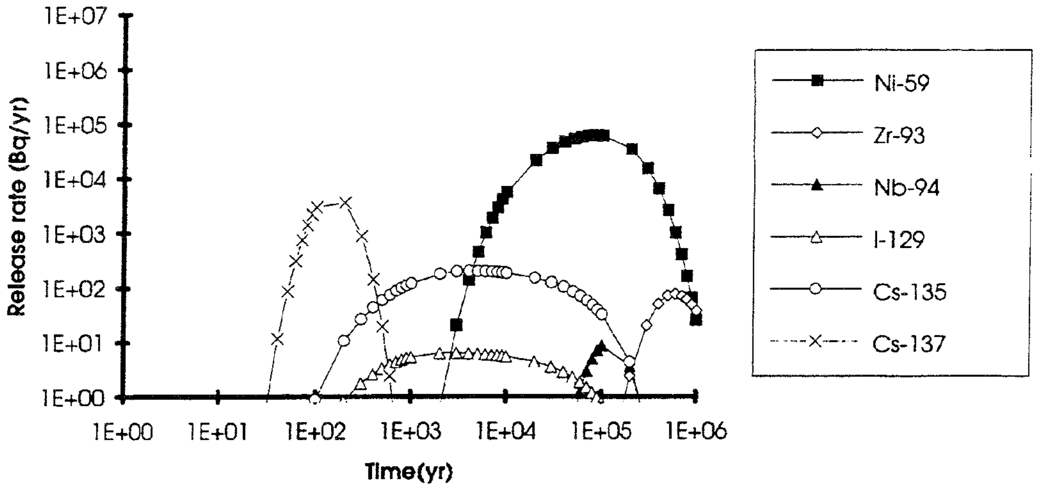

Figure 10.6 Radionuclide release from SFL 5, Reference Case. The release of Ni-59 is solubility limited and the release of Zr-93 and Nb-94 corrosion limited.

Table 10.5 Maximum release rates of 1 Bq/year and above from SFL 5, Reference Case.

Radionuclide	**Half-life (year)**	**Time for max release rate (year)**	**Maximum release rate (Bq/year)**
Ni-59[1)]	$7.5 \cdot 10^4$	$8 \cdot 10^4$	$6 \cdot 10^4$
Zr-93[2)]	$1.5 \cdot 10^6$	$1 \cdot 10^6$	$8 \cdot 10^1$
Nb-94[2)]	$2.0 \cdot 10^4$	$1 \cdot 10^5$	9
I-129	$1.6 \cdot 10^7$	$3 \cdot 10^3$	6
Cs-135	$2.3 \cdot 10^6$	$4 \cdot 10^3$	$2 \cdot 10^2$
Cs-137	$3.0 \cdot 10^1$	$2 \cdot 10^2$	$4 \cdot 10^3$

[1)] *release is limited by solubility*
[2)] *release is limited by corrosion*

10.3.5 Total radionuclide release from the near-field

The total release rates from all three repository parts of the radionuclides considered in the Reference Case calculations are shown in Figure 10.7 as a function of time. During the whole simulation period the release rate is dominated by radionuclides released from SFL 4. During the first hundreds of years the release of Cs-137 from SFL 4 dominates. However, it should once again be pointed out that the selection of cesium distribution coefficient in the calculations of the release from SFL 4 may be too pessimistic. At longer times, the release of Ni-63 and Ni-59 from SFL 4 dominates.

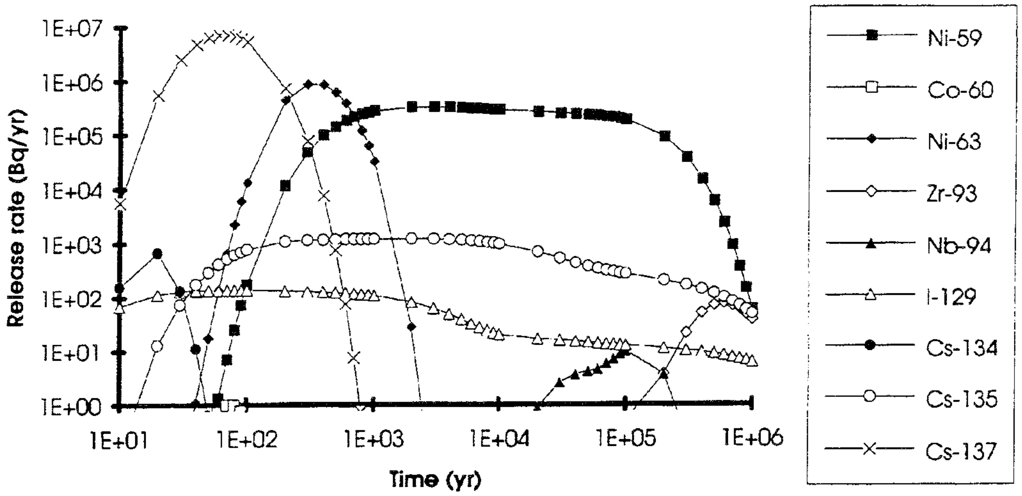

Figure 10.7 Radionuclide release from SFL 3-5, Reference Case.

Some of the radionuclides not included in the Reference Case calculations may contribute to the total release. Sr-90 will probably obtain a release curve similar to the release curve for Cs-137, with a dominating contribution from SFL 4. The maximum release rate of Ra-226 will probably not exceed half the maximum release rate of Ni-59, and Tc-99 may reach a level about one order of magnitude lower than the maximum release rate of Ni-59. Other radionuclides that may contribute to the release at early times are H-3 and C-14.

10.4 Conversion of near-field release to intermediate dose

To get a measure of the radiotoxicity of the release from the near-field of SFL 3-5, the release rates calculated for the Reference Case were converted into dose. This was done by assuming that the total release from the near-field of the repository is captured in a well with a capacity of 2000 m^3/year, and that an individual consumes 0.6 m^3 of this well water annually. This intake of radionuclides was converted to individual dose by using the dose conversion factors for ingestion given in Table 3.8. Since the effects of radionuclide retention in the far-field and the biosphere is not at all considered, the doses obtained in this way should be seen as intermediate doses, i.e. only a measure of the radiotoxicity of the near-field release.

The maximum intermediate doses corresponding to the calculated maximum release rates in the Reference Case are given in Table 10.6, and the intermediate doses as a function of time in Figures 10.8 - 10.11.

Table 10.6 and Figure 10.8 show that the intermediate dose from the SFL 3 near-field release does not exceed 10^{-8} Sv/year. The maximum intermediate dose is obtained after 100 000 years, and the major contribution arises from the release of Ni-59.

The intermediate dose from the SFL 4 near-field release are dominated by Cs-137 during early times (Table 10.6 and Figure 10.9). The maximum intermediate dose is calculated to be $3 \cdot 10^{-5}$ Sv/year, and is obtained 70 years after repository closure. At longer times, the release of Ni-63 and Ni-59 gives the highest intermediate doses.

The near-field release from SFL 5 and corresponding intermediate dose during early times is, likewise to SFL 4, dominated by Cs-137, but at a lower maximum level (Table 10.6 and Figure 10.10). The maximum intermediate dose is calculated to be 10^{-8} Sv/year and is obtained 200 years after repository closure. In the time interval 1000 to 10 000 years after repository closure the dominating contributions to the intermediate dose are from I-129 and Cs-135, and in the later parts also from Ni-59 which continues to dominate at times longer than 10 000 years.

The total intermediate doses from all three repository parts from the near-field release of the radionuclides considered in the Reference Case calculations are shown in Figure 10.11 as a function of time. From this figure and Table 10.6 it could be seen that the maximum intermediate dose during the first hundreds of years arises from the release of Cs-137 from SFL 4. However, it should once again be pointed out that the selection of cesium distribution coefficient in the calculation of the release from SFL 4 may be too pessimistic and consequently the release and corresponding intermediate dose over-estimated. At longer times, the near-field release of Ni-63 and Ni-59 from SFL 4 will give the highest intermediate doses. Other radionuclides which will contribute to the intermediate dose at longer times are I-129 and Cs-135 from SFL 4, but also from SFL 3 and SFL 5.

Table 10.6 Maximum intermediate doses from maximum near-field releases above 1 Bq/year, Reference Case.

Radionuclide	**Half-life (year)**	**Maximum intermediate dose (Sv/year)**		
		SFL 3	**SFL 4**	**SFL 5**
Ni-59	$7.5 \cdot 10^{4}$	$1 \cdot 10^{-9}$	$7 \cdot 10^{-9}$	$1 \cdot 10^{-9}$
Co-60	5.3		$3 \cdot 10^{-11}$	
Ni-63	$9.6 \cdot 10^{1}$		$5 \cdot 10^{-8}$	
Zr-93	$1.5 \cdot 10^{6}$		$6 \cdot 10^{-13}$	$3 \cdot 10^{-11}$
Nb-94	$2.0 \cdot 10^{4}$		$3 \cdot 10^{-12}$	$6 \cdot 10^{-12}$
I-129	$1.6 \cdot 10^{7}$	$4 \cdot 10^{-10}$	$5 \cdot 10^{-9}$	$2 \cdot 10^{-10}$
Cs-134	2.1		$4 \cdot 10^{-9}$	
Cs-135	$2.3 \cdot 10^{6}$	$2 \cdot 10^{-10}$	$6 \cdot 10^{-10}$	$1 \cdot 10^{-10}$
Cs-137	$3.0 \cdot 10^{1}$		$3 \cdot 10^{-5}$	$1 \cdot 10^{-8}$

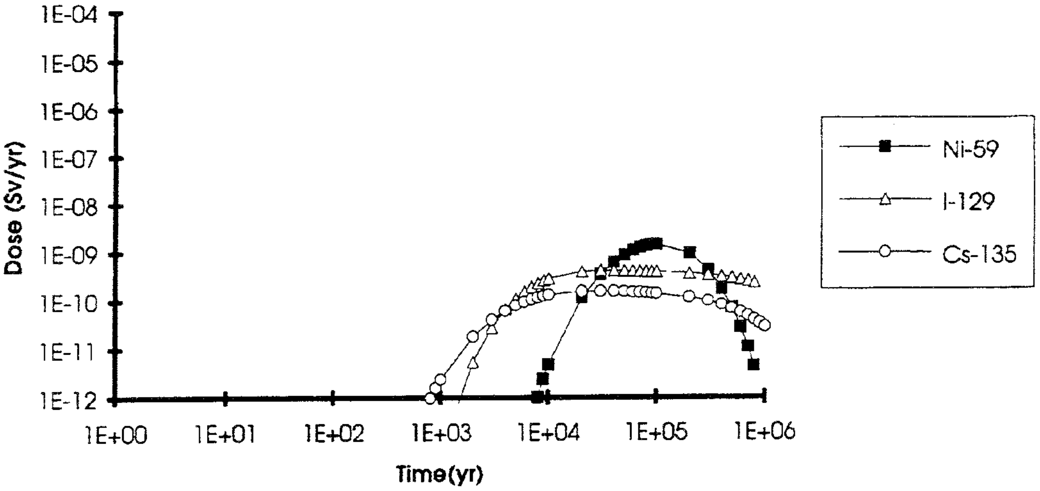

Figure 10.8 Intermediate dose from the SFL 3 near-field release, Reference Case.

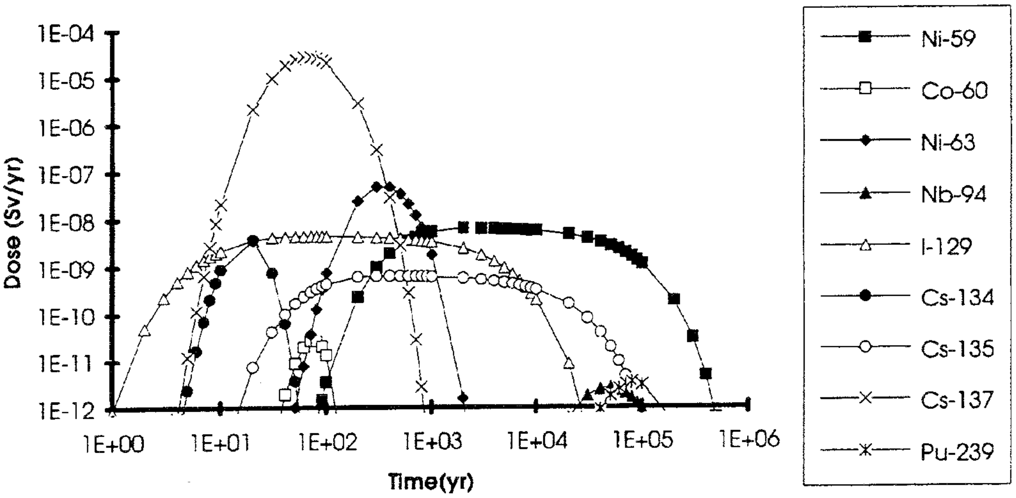

Figure 10.9 Intermediate dose from the SFL 4 near-field release, Reference Case.

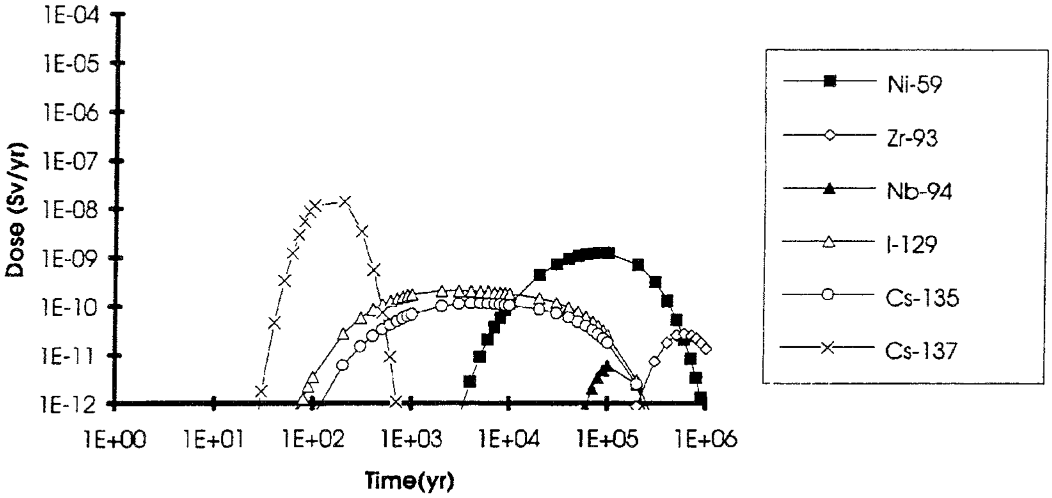

Figure 10.10 Intermediate dose from the SFL 5 near-field release, Reference Case.

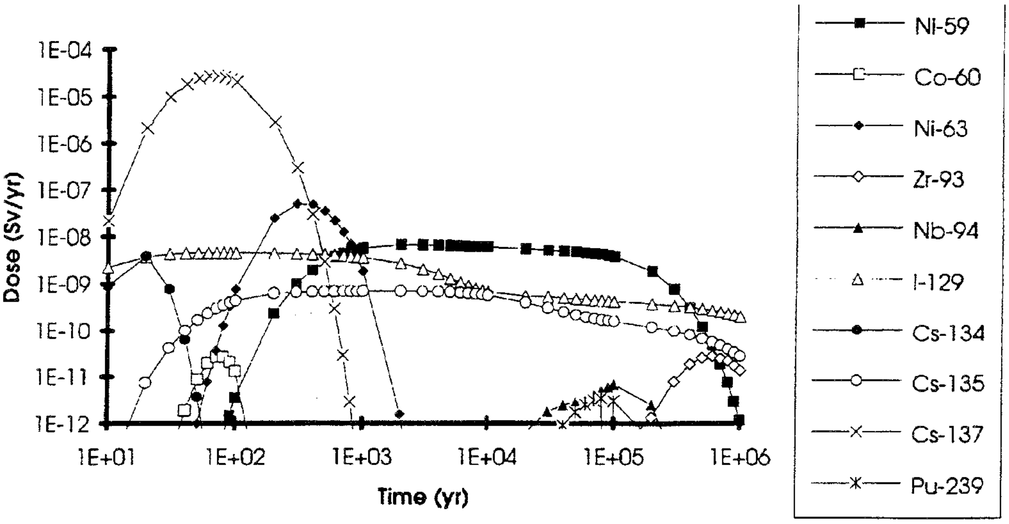

Figure 10.11 Intermediate dose from the SFL 3-5 near-field release, Reference Case.

There are some radionuclides not included in the Reference Case calculations which may obtain near-field release rates of such a magnitude that they would contribute to the intermediate dose. One example is the release of Sr-90 from SFL 4. Since the maximum release rate of Sr-90 is similar to the maximum release rate of Cs-137 (see section 10.3.3), and the difference in dose conversion factors is small (see Table 3.8), the release of Sr-90 from SFL 4 will give an intermediate dose similar to the intermediate dose from Cs-137. Adding the intermediate dose from Sr-90 will probably not result in a total intermediate dose that exceeds 0.1 mSv/year, even with the lowest of the distribution coefficients given in Table 7.1.

Another nuclide of potential importance is Ra-226 released from SFL 3. The maximum release rate of Ra-226 will probably not exceed the value calculated by the "stirred tank" model (see section 10.3.2). The intermediate dose from release of Ra-226 from SFL 3 would then not be higher than $4 \cdot 10^{-6}$ Sv/year, i.e. about 10 times lower than the intermediate dose from the release of Cs-137 from SFL 4.

Other radionuclides that may contribute to the intermediate dose at early times are H-3 and organic C-14. The effects of the release of these nuclides on total intermediate dose cannot be quantified at present.

Of the long-lived radionuclides not included in the Reference Case calculations, only the release of Th-232 from SFL 3 and the release of Tc-99 from SFL 5 may contribute to the total intermediate dose at longer times. The maximum release rates calculated with the "stirred tank" model would for both Th-232 and Tc-99 give intermediate doses of 10^{-9} Sv/year.

10.5 Release of chemotoxic elements from SFL 3

The waste in SFL 3 and SFL 5 contains elements which are classified as chemotoxic. Lead and beryllium were chosen as model substances and included as an example in the calculations of the release from SFL 3 in the Reference Case. The amount of lead in SFL 3 is estimated to 7.5 tonnes and the estimated amount of beryllium is 320 kg (see Table 3.5).

The same modelling approach was used as in the calculations of the release of radionuclides from SFL 3 in the Reference Case (see section 10.3.1). Effective diffusivities, porosities and densities used in the calculations are given in Table 4.4. Due to the slow leaching of the concrete (see Chapter 4) the lowest values of the distribution coefficients representative for fresh and leached concrete given in Table 7.1 were chosen. The distribution coefficients for bentonite and sand/bentonite were calculated from the apparent and effective diffusivities given in Table 7.2.

The calculated maximum release rate of beryllium is 20 mg/year and is obtained 400 000 years after repository closure. The release rate of lead is still increasing at the final prediction time which is 1 million years. The release rate of lead at 1 million years after repository closure is calculated to be 50 mg/year.

Assuming that the calculated maximum near-field release of beryllium and lead is captured in a well with a capacity of 2000 m^3/year would result in a beryllium concentration of 0.01 μg/l and in a lead concentration of 0.025 μg/l in the water in the well. These concentrations are well below the guideline values for drinking water which are 10 μg/l for lead [*SLV FS, 1993*] and 1 μg/l for beryllium [*U.S. EPA, 1992*].

11 Discussion

As a brief introduction, to the following discussions on waste inventory, evaluation of long-term performance, remaining issues, experimental program and comparison with SFR, major characteristics of the three repository parts SFL 3, 4 and 5 are compiled in Table 11.1.

Table 11.1 Summary of major characteristics of SFL 3-5.

Characteristics	**SFL 3**	**SFL 4**	**SFL 5**
Total waste volumes	5000 m^3	10 000 m^3	10 000 m^3
Waste origin and volumes	L/ILW Studsvik 1500 m^3 Op. waste CLAB EP 3500 m^3	CLAB cassettes 6600 m^3 Dec. waste CLAB EP 2200 m^3 Trsp. containers 1200 m^3	Core comp. Internal parts PWR 2000 m^3 BWR 8000 m^3
Packaging	Concrete mould steel drum	Steel container	Concrete container
Activity content year 2040	$3 \cdot 10^{15}$	$2 \cdot 10^{14}$	$1 \cdot 10^{17}$
Dominating radionuclides	H-3 Ni-63 Co-60 Cs-137 Pu-239 Pu-241 Am-241	Fe-55 Ni-63 Co-60 Sb-125 Cs-137	Ni-63 Co-60 H-3 Ni-59 Fe-55 C-14
Repository barriers	Concr. backfill Concr. structures Bentonite buffer	Sand backfill	Concr. structures Sand backfill

11.1 Waste inventory

Within the prestudy for SFL 3-5 a first inventory has been performed of the waste foreseen to be allocated to the repository. The uncertainties in the estimates of waste composition and radionuclide inventory are large especially regarding old waste from Studsvik. However, more information will be available during the coming years for example from ongoing projects on reconditioning and documentation of old waste at Studsvik and with the successive preparation of Waste Type Descriptions (WTD).

Material composition of waste and waste packagings

The composition and total amounts of the materials in the waste were previously known with varying degree of certainty. The waste foreseen from the nuclear power reactors and CLAB were relatively well known compared to for example research waste presently being stored at Studsvik. This made it difficult to discuss the importance of different constituents and to decide what should be concentrated on for assessment of barrier functions. The status are now much better with reasonable estimates on the total quantities of metals, organics, concrete etc, destined to the different parts in the SFL 3-5 repository.

Complexing agents in the waste can potentially enhance the release of contaminants by decreasing sorption abilities and increasing solubilities. Organic material and cyanide precipitates are examples of potential sources of complexing agents. It has been established that waste containing organic material will be concentrated to SFL 3 and that the cellulose content will be small. About 0.5% of the total weight of concrete used for conditioning and in the waste packages in SFL 3 is cellulose. Cyanide precipitates are being generated at Studsvik in water cleaning operations and appear as waste sludge conditioned with cement. This waste is also foreseen to be allocated to SFL 3. In the Swiss safety assessment equal importance were given to cyanides and cellulose as potential sources for generating mobilising complexants [*NAGRA, 1994*]. Following the same line of reasoning it could be concluded that the cyanide fraction in SFL 3 is relatively unimportant, as it only constitutes about 0.04% of the total weight of concrete in the waste packages (compared to 0.5% for cellulose). However, it can´t be excluded that the content of these potentially sources of complexing agents are concentrated to some waste packages and influences the releases from a minor part of the waste.

Steel will be present in all repository parts, as waste, waste packaging and as reinforcement in concrete containers and structures. Much of the steel in the waste is stainless steel. Other metals and metal alloys present are aluminium, Zircaloy, lead, brass, copper, cadmium, etc. The quantities of these metals are minor relative to the total content of steel in the repository (less than 1%). Aluminium, which is a potentially strong gas generation source is concentrated to SFL 3. The total estimated quantity of aluminium in SFL 3 is about 51 metric tonnes.

Concrete/cement will be present in all repository parts. SFL 3 and SFL 5 will contain large amounts of concrete/cement either as waste conditioning material or waste backfill, but also as waste container material. In addition, concrete will be used for the underground structures and in SFL 3 also as backfill material. However, SFL 4 will contain relatively small amounts of concrete. Some concrete debris from decommissioning operations is destined for SFL 4, but apart from that there is at present no concrete structures intended in SFL 4, except for paving and shotcrete. So far only steel containers have been considered as packagings in SFL 4.

Radionuclide content

It should be remembered that the radionuclide content in the waste is based on estimates. For example, CRUD contents have in most cases been calculated from surface areas and information on contamination levels on various materials which have experienced an exposure situation similar to the waste destined to SFL 4 and 5. The relation between CRUD and induced activity is a bit uncertain. There are also differences in accuracy in the information from different sources. For example, waste from BWRs is at present better covered than waste from PWRs.

However, it is quite clear that SFL 5 and its content of induced activity in the metallic waste from the reactors will determine the total activity in the repository SFL 3-5 at repository closure. The two Ni-isotopes Ni-59 and Ni-63 will dominate, Ni-63 during the first 1000 years and Ni-59 thereafter.

SFL 3 will according to first estimates contain more than one order of magnitude less total activity than SFL 5. The dominating nuclides initially are H-3 and Ni-63. The tritium is concentrated to one waste category which is H-3 in titanium hydride. After 1000 years Ni-59 will dominate the activity in the SFL 3 waste.

The total activity in SFL 4 will always be more than one order of magnitude less than in SFL 3. The nuclides Fe-55 and Co-60 will dominate during the first 20 years. Thereafter, Ni-63 is the dominating nuclide until 1000 years after repository closure. After 1000 years Ni-59 will dominate the activity.

The contribution to total activity is not enough to judge the importance of different radionuclides for the long term safety. The dominating nickel isotopes have relatively low radiotoxicities compared to for example Cs- and Pu-isotopes, which are found in the largest amounts in the SFL 3 waste. In addition to the radiotoxicity the availability of the radionuclides in the waste and their retention in the barriers must be considered. For example, the induced activity is the main source of nickel isotopes and they are thereby well protected in the stainless steel components and, moreover, concrete is a good barrier for nickel.

Chemotoxic components

Not only the radionuclide content, but also the content of potential chemotoxic elements must be considered in a safety assessment. SFL 3 will according to first estimates contain chemotoxic elements in some waste types of noticeable quantities, for example certain metals like cadmium (1.2 tonnes), lead (7.5 tonnes), beryllium (0.3 tonnes) and uranium in ashes (2.9 tonnes). The waste in SFL 5 contains lead, which has been used for shielding of the TIP detectors (124 tonnes), and beryllium from decommissioning of the research reactor (R2) at Studsvik. However, it may be possible to dispose the TIP detectors without the lead shielding. In SFL 4 the waste comprises mainly stainless steel components, and it is questionable if the content of chromium and nickel in the steel should be regarded as chemotoxic components.

11.2 Evaluation of long-term performance

In the prestudy a first evaluation of the long term performance of the SFL 3-5 repository has been made. The methodology used to formulate the studied Reference Scenario and the processes and mechanisms considered in the analyses of the near-field release are briefly discussed in this section.

Scenario development

A systematic methodology for scenario formulation was tested. It was possible to carry through the different steps in the methodology, to formulate a Reference Scenario for SFL 3-5 and to define a Reference Case to be quantitatively analysed. An important result from this part of the study is the data base with documents, protocols and decisions behind the development of the Reference Scenario. This will facilitate future re-evaluations.

The approach of constructing Influence Diagrams was used in order to structure graphically the Features, Events and Processes, FEPs, within the so called Process System. An advantage with using Influence Diagrams is the possibility to schematically represent the actual lay-out of the repository system. A drawback is the complex system of boxes and arrows, which is a consequence of illustrating all phenomena and their interactions involved in mobilisation and not only the transport paths through the system. Finding a way to graphically structure the Process System, and thereby make it more transparent without loosing information, would improve the methodology.

It should be noted that studies of this kind is not a way to obtain completeness, in the sense that all possible scenarios have been identified. However, we have found this to be a feasible way to compile, treat and document a very large number of issues of potential importance for the long-term performance of the repository.

Water flow in the repository

The groundwater flow in the repository has been estimated with an analytical model and is based on generic site conditions. The maximum water flow in the SFL 4 and SFL 5 is a few m^3 annually when the repository barriers are considered to not limit the water flow and the average water flux in the rock is assumed to be 0.1 l/m^2,year. In SFL 3 the bentonite barriers are assumed to restrict the water flow and the contaminant release is governed by diffusion.

Chemical stability in the repository

Concrete has a strong and mainly beneficial influence on the chemical conditions in SFL 3 and 5. The laboratory experiments and calculations performed were valuable by proving the long-term stability of these conditions. We therefore assumed, that pH was above 12.5

for at least 900 exchanges of pore water. Due to the slow flow of groundwater in the repository, it was not necessary to consider a more leached and degraded concrete than that and pH was therefore considered to remain high for up to 10^6 years. The high pH is an advantage because it has a generally positive effect on the sorption except for cesium and strontium, which sorb by ion-exchange and must compete with the calcium in the concrete porewater.

Gas formation

Gas can in principle be generated in the repository by radiolysis, microbial degradation and corrosion. Gas formation by radiolysis, microbial degradation and corrosion of metals in the waste and the waste packagings was considered in the prestudy. The annual gas formation by radiolysis was estimated to be only a few m^3(STP) in the most radioactive repository part SFL 5. The initial gas formation rate by microbial degradation of cellulose and other organics, mainly ion-exchange resins, in the SFL 3 waste was estimated to about 50 m^3(STP) and will decrease to a few m^3(STP) annually when the cellulose is degraded. The gas formation by corrosion of metals was found to be initially determined by the content of aluminum in the SFL 3 waste, whereas the potential total amounts of gas formed are determined by the steel content. After the aluminium is depleted the gas formation due to corrosion of steel was estimated to about 2000 m^3(STP) annually. The aluminium in the SFL 3 waste originates mainly from ILW from Studsvik and the amounts can be overpredicted. In the estimates of gas formation the contribution from the reinforcements in the concrete structures was not included.

Temperature

The increase in temperature, due to radioactive decay of nuclides, in the waste and the repository has been evaluated. The increase in temperature in the most active repository part SFL 5 was estimated to be only a few degrees.

Calculated near-field release of radionuclides

A Reference Case was defined and the near-field release of radionuclides was calculated. The conclusions from this preliminary exercise should not be carried too far, but it deserves to be mentioned that very low release rates were obtained.

The nuclides selected for the assessment of barrier functions were Ni-59, Ni-63, Co-60, Zr-93, Nb-93m, Nb-94, I-129, Cs-134, Cs-135, Cs-137, Pu-238, Pu-239, Pu-240, Pu-241 and Am-241. The selected radionuclides were judged to be representative with respect to relative importance concerning abundance, radiotoxicity and mobility. For example, the nickel isotopes are very abundant in the waste, iodine is very mobile, and cesium and plutonium have relatively high dose conversion factors.

The calculations revealed that among the selected radionuclides Cs-137 and Ni-63 would dominate the annual release from all repository parts during the first 1000 years after repository closure and that Ni-59 would dominate at longer times.

The highest release rates arises from SFL 4, despite the fact that the total content of nuclides are more than one order of magnitude less compared to the other repository parts. This is partly explained by the conservative CRUD estimates and partly due to the fact that the only barrier in SFL 4 considered in the calculations is the sand-backfill. The main reason is the limited retention and the water flow in the sand-backfill.

Conversion of near-field releases to intermediate doses

Intermediate doses calculated from the near-field radionuclide releases were always below 0.1 mSv/a for a simplified well scenario. The dose dominating radionuclide during the first 1000 years is Cs-137 and at longer times Ni-59.

Near-field release of chemotoxic elements

Lead and beryllium were chosen as model substances and included as an example in the release calculations of the Reference Case for SFL 3. For the same simplified well scenario as for the radionuclide release it was found that calculated near-field releases of lead and beryllium were below the guideline values used for drinking water. This demonstrates that some of the barriers effective to prevent release of radionuclides, such as sorption in concrete, are also efficient in this case.

11.3 Remaining issues

Identified aspects that have to be considered in future evaluations of the long-term performance of the SFL 3-5 repository are discussed in this section.

Repository design

The evaluation of barrier functions in the prestudy has been based on a preliminary repository design of SFL 3-5. Already in the prestudy questions have been identified that can have implications on the development of a more detailed barrier and repository design. One example is the selection of barrier material in the different repository parts.

Another example concerns the function of the shafts and/or tunnels that connect the repository with the surface. To isolate the waste from the environment a successful sealing of the shafts, tunnels and boreholes is essential. In case of "rupture" of the seals or a less successful sealing the groundwater flow through the repository might increase significantly. The influence of different lay-out/location of shafts should be evaluated. In particular, a cross connection between shafts on both sides of the repository.

Reference Scenario

Processes and mechanisms assessed to be of potential importance for the performance of the repository, but which have not been considered in the analyses of the Reference Scenario within the prestudy are discussed in Section 6.3.2 and summarised below.

- Processes directly involved in the mobilisation and out-transport of radionuclides and chemotoxic elements which so far have not been considered in the calculations are e.g.gas generation and transport, sorption/coprecipitation with corrosion products, coprecipitation with calcite and colloid generation and transport.
- The different near-field barriers in the proposed repository design may in the long-term perspective be altered and even degraded due to interaction and chemical reaction with the water and its constituents and due to mechanical impact. The only alteration/degradation process considered in the prestudy analyses of the Reference Scenario is leaching of cementitious components from the concrete barriers in SFL 5.
- Despite the short duration of the resaturation phase in relation to the time of interest of repository performance, there are some aspects of the resaturation phase that may be of importance.

Other scenarios

In addition to the defined Reference Scenario other scenarios describing the potential influence of initial conditions in the repository and the influence of altered environmental conditions may have to be studied to evaluate the long-term performance of the repository. Potential scenarios are:

- Scenarios covering changes of the environmental conditions, for example climatic changes at the repository site. Altered environmental conditions may influence the groundwater flow and direction, the groundwater composition and thereby the function and properties of the barriers in long-term perspectives.
- Scenarios covering different initial conditions in the repository, for example the operation of the repository may alter the expected chemical conditions in the near-field barriers as well as the temperature compared with undisturbed rock at the same levels.
- In the Reference Scenario the repository parts are assumed to be isolated from each other. However, other scenarios considering possible interactions between SFL 3, 4 and 5 may have to be studied, for example the influence of complexing agents and organic material in SFL 3 on the performance of the other repository parts, the distribution and interaction of water flow between different repository parts, etc.

Interactions between SFL 2 and SFL 3-5

An example of other scenarios that has to be included in future safety assessment is the interactions between SFL 2 and SFL 3-5. The repositories SFL 2 and SFL 3-5 are planned to be located adjacent to each other and to be accessed from above by the same service facility. The repository areas are foreseen to be separated by a 1 km long transport tunnel and to be isolated from each other by appropriate plugging and backfilling of the tunnel systems. However, in long term perspectives it cannot be excluded that the existence of one of the repositories may influence the performance of the other repository.

The SFL 2 repository performance relies mainly on the canister integrity and protection by the surrounding bentonite buffer. Important barriers in the repository for spent nuclear fuel, SFL 2, are the waste canister (copper and steel) and the compacted sodium bentonite. The low solubility of the spent fuel matrix and the low solubility, low diffusivity and high sorption of many important radionuclides are important processes that delay the release and dispersion of radionuclides in case of canister failure.

The SFL 3-5 repository performance relies mainly on the unavailability and immobility of the radionuclides. Sorption and retardation in the engineered barriers and a limited water flow through the repository are important for its performance.

Some topics that may influence the long-term performance of SFL 2 and SFL 3-5 which have to be studied are discussed below.

- In SFL 2 located adjacent to SFL 3-5 heat is generated. The temperature is reported to reach a maximum of about 70 °C on the canister surface after ten years and then decrease. This increase in temperature is estimated to not appreciable affect the water flow in SFL 2 or the SFL 3-5 barriers.

- Corrosive agents and aggressive species from the waste and the engineered barriers in SFL 3-5 can influence the canister life time and the stability of the bentonite buffer. According to the present concept SFL 2 will be separated from SFL 3-5 by a distance of about 1 km. It is difficult to imagine that any release of constituents would arrive in any significant concentration after a travel distance of 1 km horizontally. The travel time will be considerable, dilution will take place and reactions along the flow path can be envisaged, which consumes the released compounds. Let us for example assume a release of hydroxide and potassium ions from the concrete in SFL 3-5. None of these constituents are wanted in SFL 2. However, the pH of the groundwater is stabilised by reactions with components in the water (bicarbonate) and the rock minerals. Silicates in normal granite become reactive to solutions when pH rises above 11. Potassium will react with the rock minerals and that will reduce the concentration of potassium. This can be observed in places where sea water has infiltrated the rock.

- The metals contained in SFL 3-5 in the form of concrete reinforcement (steel), waste containers (steel) and scrap metals (steel, stainless steels, Zircaloy, aluminium etc) will corrode and generate hydrogen gas. Hydrogen gas formed can, at least in principle, be transported to SFL 2 as gas or dissolved in the water. Hydrogen can potentially be used by microbes [*Pedersen, 1993*]. It deserves some consideration because in principle it could constitute a route for sulphate reduction to sulphide which can act as a corrodent for iron and copper.

- Degradation products from cellulose in SFL 3 can have a negative influence on radionuclide release and retention by increasing the solubility and lowering the sorption. However, these compounds known as polyhydroxo-carboxylic acids are only effective at very high pH-values (at pH 12 and above). Dilution and pH-adjustment will make them less important away from the concrete dominated environment in SFL 3-5. These compounds are also known to be easily decomposed by bacteria, which is another reason why they should become reduced.

- It should be feasible to place the repository SFL 3-5 downstream SFL 2 relative to the horizontal component of the predominant groundwater gradient. This would be a efficient way to avoid the possibility of any future interaction altogether. Another way would be to locate the repositories with a water divide in between them. These and other possibilities to adapt the repositories to the hydraulic conditions at a site have not been discussed at any depth within the prestudy. However, it is probably worthwhile to consider this in the future when the lay-out of the repositories at a particular site is being selected.

11.4 Experimental program

It will be necessary to analyze the long-term safety for disposal of this waste as have already been done for spent fuel [*KBS-3, 1983, SKB 91, 1992*] and short-lived LLW and ILW [*Carlsson et al., 1991*]. The work on a full safety assessment may begin by mid 1996 and the preparation of that exercise is an important task for the SKB program on long-lived LLW and ILW. This is specifically stated in one of the three general aims; "to prepare the safety assessment and gather data that will become necessary at a later stage". However, when this program was started at the end of 1992, we realised that any data that had to be measured in the laboratory would take us time to get. It would be important to start laboratory work early. On the other hand it is very difficult to know exactly how important a certain parameter value is until it has been used in a performance assessment. The Prestudy Project may be seen as an attempt to solve this dilemma. The aim of the prestudy is; "to make a first preliminary and simplified assessment of the near-field barriers to radionuclide dispersion". Experimentalists and safety assessment specialists have been engaged together in this project and the intentions have been to stimulate development and direct the efforts to the important properties of the barriers.

Radionuclide chemistry

The retention of radionuclides in the concrete dominated environment has turned out to be an important barrier function and the following experiments were started in order to determine how the radionuclides would react in groundwater and cement in a repository:

- Sorption of Eu, Th, Np, Am, Cm, Pm, Co, Ra, Ni and Cs in concrete.

- Diffusion of Ni, Cs and T in the cement paste.

- Solubility of Ni, Pu and Eu in cement porewater.

It should be noted that not all of the elements have been chosen because they appear as important radionuclides in the waste. Some have been selected because they have chemical properties we want to study, for example Eu, Cm and Pm which are typical +III-valent elements with suitable radioisotopes for the experiments (half-life and radiation decay).

The actinides Th, U, Np and Pu are all expected to appear in their tetravalent state in the waste due to the redox conditions there. It has been argued that the tetravalent actinides should form negatively charged hydroxy complexes at the high pH-conditions prevailing in concrete and that the existence of such complexes could increase solubility and decrease sorption retention. However, the experiments made support the conclusions drawn already before the prestudy that there are no such negative complexes for the tetravalent actinides [*Engkvist, 1993*]. Their mobility will be very low indeed unless they are affected by some other strongly complexing agents. Cellulose degradation products may be an example (see below).

Bentonite has been suggested as a backfill material between the rock walls and the inner concrete structures in SFL 3. Radionuclide diffusion in pure bentonite is rather well studied, but there was a lack of data for the backfill in the top and bottom of SFL 3 which are composed of sand/bentonite mixtures. Therefore the following radionuclide diffusion experiments were added:

- Diffusion of Cs, Tc and Ni in a sand/bentonite mixture (85/15).

The results have not been published yet and in fact many of the radionuclide chemistry studies that were initiated are still going on. However, preliminary results have been used in the prestudy and guidance have been found in the selection of values to use as input to the calculations.

Concrete stability

Concrete has important barrier functions in SFL 3 and 5, which are the parts that contain most of the radionuclides. It is therefore important that concrete retains it properties in the long-term perspective. We may of course have to accept a certain mechanical damage to the construction structures, packages etc during the enormous time-spans considered. However, in particular the chemical stability of concrete is important, because concrete porewater will in general enhance retention and suppress solubility of radionuclides in the waste. It is necessary to know if we should expect any significant change in the time period of interest which needs to be included in the calculation of radionuclide dispersion.

Two efforts have been directed towards this aim; one experimental investigation and one study mainly of published results:

- Static leaching of cement paste.
- Summary of the knowledge on concrete degradation in a deep underground environment.

The last mentioned study did also include the examination of an old (90 years) concrete sample from a fresh water tank in Uppsala. The overview report will be published in the SKB technical report series [*Lagerblad and Trädgård, 1995*].

The investigations of concrete durability have strengthened our conviction that the typical concrete dominated high pH-conditions will remain in the repository for as long as we need to consider the hazards of the waste. It should not be necessary to consider any scenario with $pH < 12$, at least not in SFL 3 and 5. However, in SFL 4 there is not enough concrete to guarantee such conditions and it is therefore necessary to assume a range of possibilities in that particular part of the repository; from pure groundwater to concrete leachate.

Cellulose degradation

It has been observed, first by researchers at Harwell Laboratories in England [*Greenfield et al., 1991*], that cellulose in concrete has an influence on the chemistry of radionuclides. It was for example demonstrated by experiments that cellulose products considerably increased the solubility of plutonium in concrete porewater. A possible explanation for these observations is that cellulose is hydrolysed at high pH and that the new compounds formed as a result of the hydrolysis are strong complexing agents. The expected products are so called polyhydroxo-carboxylic acids. Examples of such compounds are D-gluconic acid and D-glucoisosaccharinic acid.

In order to shed light on these conditions and to find ways to quantify the potential influence of cellulose degradation on radionuclide chemistry the following experimental activities were initiated:

- Hydrolysis of cellulose at high pH.

- Chemical degradation of gluconic acid at high pH.

- Formation of metal complexes with polyhydroxo compounds.

- Influence of polyhydroxo-carboxylic acids on the sorption of metal ions in concrete.

Results so far have been summarised by Maria Nordén in her Doctoral Thesis [*Nordén, 1994*]. Considering this and other recently published results [*Baston et al., 1994*] we conclude that:

- The occurrence of cellulose degradation will enhance the solubility of particularly tetravalent elements at high pH.

- The major potential complexing acid that can be formed at substantial yield by alkaline degradation of cellulose is D-glucoisosaccharinic acid.

- The presence of cellulose degradation products at levels of 20-30 mg/l or above would lead to an initial reduction of the absorption of tri- and tetravalent elements on mineral surfaces at high pH such as cement at pH 12.

- The presence of the acidic hexose derivatives D-gluconic acid at concentrations above 10^{-4} M and D-glucoisosaccharinic acid (at slightly higher concentrations) would lead to an initial reduction of the adsorption, as observed in real mixtures of cellulose degradation products.

- The reduction of adsorption due to the formation of complexes with hexose derivatives etc cannot be calculated from the corresponding increased solubility; evidently the complexes are to a significant extent sorbed on the solid surfaces as well.

- Initial effects on metal ion adsorption due to gluconic or isosaccharinic acids are decreasing with time at pH above 12, particularly for isosaccharinic acid. The effect on sorption is only minor after 5-6 months (gluconic acid) and 1-2 months (isosaccharinic acid) at initial concentrations in the mM-range.

It is worth to notice that only SFL 3 contains cellulose in quantities that are of any importance ($<0.5\%$ of the weight of concrete in the waste packages).

11.5 A comparison with SFR

SFL 3-5 has many similarities with the final repository for radioactive operational waste, SFR 1, in operation and the final repository for decommissioning waste, SFR 3, planned to be built at the same location. The waste is similar and many cases even identical, for example the filter masses from CLAB. Also the design is based on similar principles; excavated rock vaults barriers of concrete and different backfill materials, such as concrete, gravel and bentonite clay. SFL 5 is similar to the BMA vault in SFR and SFL 3 have many barrier features in common with the silo.The waste streams are allocated to different parts of the repositories depending on the origin, activity content and chemical properties of the waste. The time of production is of some importance, such that waste packages produced after the closure of SFR 1 and SFR 3 will be allocated to SFL.

Design

SFR is mainly designed for short lived LLW and ILW from the operation of the Swedish nuclear power plants. Later it will also receive waste from the decommissioning of the power plants. SFR will also receive waste from research, industry and medicine. The first constructed part, SFR 1, is now in operation and receives operational waste. The disposal vaults are caverns (length 160 m and cross section of approximately 200 m^2) of three different types and a silo (diameter 30 m and height 50 m). In the caverns (except one) the waste will be backfilled with porous materials e.g. sand, gravel or porous concrete. In the fourth cavern, for low level scrap material, no backfilling is planned. The waste disposed of in the silo is backfilled with porous concrete. The silo is surrounded by bentonite clay with low water permeability. SFR 3, is a planned extension for disposal of the short lived decommissioning waste from the power plants.

The long lived, low and intermediate level, waste repository consists of three parts, SFL 3, 4 and 5. Each part is a cavern with engineered barriers of concrete and backfill. SFL 3 has a bentonite backfill between the concrete structure and the rock. SFL 4 is just the tunnel system used for transports and excavation of SFL 3 and 5. The last part, SFL 5, has a concrete structure, but only sand as backfill.

Comparison between silo and vault

A silo has been built in the SFR, but in the SFL 3 concept a "caisson in cavern" construction has been suggested. A bentonite barrier surrounding the concrete structure has been considered in both designs. It is interesting to compare the two approaches, cavern or silo, in a more systematical way. In the following, different aspects on operation and long-term safety are discussed for the two alternatives.

From the aspect of being a mechanical support for the bentonite over very long time-periods the curved form of the silo is preferred before the flat walls in the cavern concept. A curved wall can take up the pressure from bentonite swelling and rock creep without the tensional stress that would develop in a flat wall. However, partially curved walls in the cavern might be a solution that prolongs the mechanical stability also in this concept.

The effective lifetime of the bentonite is defined as the minimum time it will take to change Na-bentonite into Ca-bentonite. According to this definition a one meter thick bentonite layer has a lifetime of 1000 years assuming the same condition as those prevailing at the SFR-silo in Forsmark. However, Ca-bentonite has the properties of swelling clay too and therefore transport governed by diffusion through the bentonite can be anticipated for much longer periods, if the bentonite is kept in place mechanically.

The degradation of concrete and corrosion of steel reinforcement bars will lead to a consolidation of the concrete construction at some time in the future. The top of a high silo will then "collapse" more than the top of a concrete structure in a tunnel. The consolidation is normally a few percent of the total height of the concrete structure. The initially low permeability of the top buffer will increase due to the consolidation because of the resulting volume expansion in the top-part.

For at least some hundreds of years a concrete construction can be strong enough to resist the swelling pressure from bentonite, provided that the bentonite is not highly compacted and that the construction is equipped with some inner supporting walls.

A prolonged lifetime of the bentonite barrier can be achieved by e.g. thicker barriers or higher degree of compaction. In both cases also higher swelling pressures will be reached. The silo is the better construction concept because it can take up swelling pressures better than flat walls.

In the case of a mixed bed with sand and bentonite, as in the bottom and top layers, the "lifetime" might be extended by increasing the bentonite part of the mixture. Increasing the bentonite also implies that the mechanical stability of the barrier is jeopardised. For a bottom bed it means that if too much bentonite is mixed with the sand the inner concrete construction (silo or caisson) will "sink" through the bed due to its weight.

The mechanical stability (stackability) of the waste packages has to be higher when disposed of in the silo than in a cavern-caisson. However, weak packages can be put in separate disposal overpacks or a relatively strong backfill (concrete) can be used in order to allow for disposal in a deep silo.

Backfilling of the silo has to be done successively during disposal in order to stabilise the bottom layers of waste packages and to guarantee a complete infilling. In the cavern on the other hand the time for backfilling can be chosen relatively freely.

The benefit of backfilling inside a silo or cavern is partly depending on the waste packages. Voids inside waste packages will influence the long term mechanical stability. Large void volumes will allow for large deformations of the structure regardless of whether empty spaces outside the packages have been backfilled. This is an important factor for the long-term stability of structures where bentonite is used as a barrier.

The equipment for remote handling of the waste will be about the same in a silo as in a cavern.

A silo has to be deep enough or else the diameter will be larger than the width of a cavern with the same storage volume. The span of the roof is of importance due to rock stability. Less span makes the roof more stable. There are two ways to decrease the roof span for a silo; to excavate a deeper silo or to build several smaller silos.

The silo concept means less excavated volume outside the disposal volume in the vault than in a cavern concept. Less volume means less water flowing around the waste and thereby also less release of radionuclides by water transport.

In gas release calculations the silo concept is preferred before a cavern concept. The reason is that an over pressure in the silo expels a smaller amount of water from a silo than from a cavern. Another advantage with the silo concept is that it is easier to demonstrate that an over pressure will result in fractures in the top rather than in the bottom of the structure.

Taking the pros and cons together for the silo and caisson-cavern alternatives it is not possible to see any clear preference. Both alternatives are possible and give a comparable level of long-term protection of the waste.

Waste

The low and intermediate level waste in SFR and in SFL 3-5 are similar in material composition and some of the waste from CLAB which is expected to be disposed of in SFL 3 will be exactly the same as what is now being disposed of in SFR 1. Other waste categories contain more long-lived radionuclides and are therefor intended for SFL rather than SFR. However, the difference is more in quantity than in kind. The same nuclides are present in the SFR-waste, but in relatively minor quantities.

As mentioned above, some of the waste disposed of in SFL 3-5 is in fact also acceptable for disposal in SFR. The reason for being deposited in SFL is that the waste arises after the closure of SFR. The waste volume in SFL 3 has been estimated to about 5000 m^3. From this volume 3700 m^3 is of the SFR 1 type i.e. more than 70% could be allocated to SFR 1. In SFL 4 the volume has been estimated to 10 000 m^3 where almost 90% is of SFR 3 type. Only SFL 5 consists of solely SFL type of waste and the estimated volume is about 10 000 m^3. In summary, approximately 50% of the waste volume in SFL 3-5 consists of waste types that also are acceptable for disposal in SFR 1 or SFR 3.

Activity content

Comparing directly the projected amounts of radionuclides in SFL 3-5 and SFR 1 at the individual times of closure for the two facilities reveals, that SFL 3-5 contains about one order of magnitude higher activity than SFR 1. The reason for that is the relatively high content of Ni-63, most of it in the form of induced activity in metallic parts located to SFL 5. However, it is interesting to note that when comparing radiotoxicity, SFL 3-5 falls below SFR 1 at the times of closure. This is due to the relatively low radiotoxicity of Ni-63 compared to for example Cs-137 which is more important in SFR. After 1000 years not only the total activity, but also the radiotoxicity of the SFL 3-5 waste dominates over the SFR 1 waste, see Figure 11.1. The difference is not dramatic, but it may warrant the deeper location of SFL 3-5.

Gas formation

Gas formation in a repository may enhance the release of potentially contaminated water from the near field to the host rock, if the gas cannot be transported away without extensive pressure build-up. This has been analysed in detail for the SFR 1 silo. In the silo approximately 4000 m^3(STP) per year of hydrogen gas is assumed to be produced. This volume is reduced to 800 m^3 at repository depth. It has been shown by calculations and experiments that this annual gas production can escape and will only marginally influence the radionuclide release.

In SFL 3-5 gas formed in SFL 4 and SFL 5 can be anticipated to be transported away without extensive pressure build-up through the repository barriers. However, in SFL 3 bentonite barriers are foreseen to surround the concrete structures. In this repository the annual production of gas has been estimated to about 20 000 m^3(STP) initially, from corrosion of aluminum waste. Due to the depth of the repository the actual volume of gas formed annually is reduced to 400 m^3. When the aluminium is consumed the annual gas formation rate will decrease to about 5 m^3, by corrosion of steel in waste and waste packagings. The contribution from other metals that can be found in repository structures, e g reinforcements, has not been considered within the prestudy.

Even though the amounts of gas annually formed is less in SFL 3 compared with the silo in SFR 1 no conclusion can be drawn on the effects and the acceptability due to differences in design.

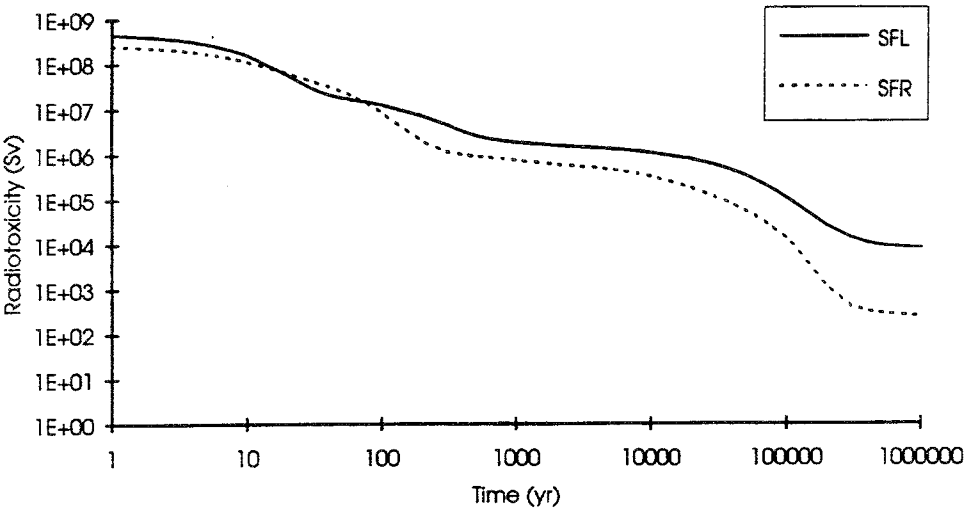

Figure 11.1 Comparison of radiotoxicity at repository closure for SFL 3-5 and SFR 1.

12 Concluding remarks

The prestudy summarised in this report has involved a first attempt to characterise the waste presently planned for SFL 3-5, the testing of a systematic scenario methodology and a first evaluation of barrier performance and containment of radionuclides and chemotoxic elements. It should be pointed out that the waste characterisation is based on rough estimates and that the evaluation of barrier performance and containment so far is restricted to the defined Reference Case which only includes parts of the identified Reference Scenario mechanisms.

The main findings from the prestudy are summarised in the following concluding remarks:

- Due to the metallic waste from the reactors, SFL 5 will dominate the total activity in the repository SFL 3-5. At repository closure, the activity in SFL 3 waste will be more than one order of magnitude less than in the SFL 5 waste, and the activity in the SFL 4 waste will always be more than one order of magnitude less than in the SFL 3 waste.

- During the first 10 000 years the radionuclide release from SFL 4 is dominating, despite that the radionuclide content in the waste in SFL 4 is more than one order of magnitude lower than in the other repository parts. The main reason for this is the limited retention in SFL 4. The dominating nuclides are Cs-137, Ni-63 and Ni-59.

- At times longer than 10 000 years, the radionuclide releases from the repository are dominated by the release from SFL 3 and SFL 5. The dominating nuclides are Ni-59, Zr-93 and Cs-135.

- Assuming that the total near-field release is captured in a well with a capacity of 2000 m^3/year and an annual water consumption of 0.6 m^3 per individual, the estimated intermediate doses are always below 0.1 mSv/year for all studied radionuclides. The highest intermediate dose arises from the release of Cs-137 after about 100 years.

- A well capturing the near-field release of the chemotoxic elements lead and beryllium from SFL 3 would obtain concentrations of these elements that are below the guideline values for drinking water. This demonstrates that barriers effective to prevent release of radionuclides are also efficient in this case.

Other findings from the prestudy are summarised below:

- It was possible to carry out the different steps in the systematic scenario methodology based on Influence Diagrams to obtain a Reference Scenario and to define a Reference Case to be quantitatively analysed as well as to prepare a list of remaining issues which should be addressed in future studies. An important result so far is the data base with FEPs descriptions, specification of influences and protocols of the decisions behind the development of the Reference Scenario. A drawback is the graphical description with its complex system of boxes and arrows.

- The waste volume including the packages is estimated to about 5000 m^3 in SFL 3, 10 000 m^3 in SFL 4 and 10 000 m^3 in SFL 5, giving a total volume of about 25 000 m^3 in the whole repository. This volume is less than one third of the waste volume that is planned to be disposed of in SFR 1 (76 100 m^3).

- The activity content in the SFL 3 waste during the first 100 years after repository closure is dominated by H-3 in titanium hydride and here after by Ni-63. The two Ni-isotopes are dominating radionuclides in the waste both for SFL 3 and SFL 5, Ni-63 during the first 1000 years after repository closure and Ni-59 thereafter. This is also the case for the SFL 4 waste except during the first 20 years when Fe-55 and Co-60 dominates the activity.

- The total activity in SFL 3-5 at repository closure will be approximately 10^{17} Bq. This is one order of magnitude higher than the activity in SFR 1 at repository closure. If the radionuclide specific dose conversion factor for ingestion is used to convert the activity content to potential doses, the total radiotoxicity of the waste in SFL 3-5 at repository closure will be approximately half of the radiotoxicity in the waste in SFR 1 at repository closure. The radiotoxicity in SFR 1 is dominated by the content of Cs-137 in the Silo and the radiotoxicity in the SFL 3-5 is determined by the content of Co-60 and Ni-63 in the SFL 5 waste. At longer times, approximately 1000 years after repository closure, the radiotoxicity is dominated by the long-lived actinides Am-241, Pu-239 and Pu-240 in the silo and the SFL 3 waste. At this time the radiotoxicity of the waste in SFR 1 has decreased more than the waste in SFL 3-5. The radiotoxicity in SFL 3-5 is now about twice the radiotoxicity in SFR 1, see Figure 11.1.

- The lowest contribution to the radionuclide releases from the repository arises from the waste in SFL 3. This is mainly due to the fact that the short-lived nuclides do not survive the diffusive transport through the concrete and bentonite barriers.

- The temperature increase due to decay of radionuclides in the waste was calculated to be less than 2 °C and is therefore neglected.

- The dominating metal waste is steel, mostly stainless steel, which will be present in all repository parts. The total quantity is estimated to about 11 000 metric tonnes. Aluminium, which has a strong potential for gas formation, is mainly found in the Studsvik waste. The waste is allocated to SFL 3 and the estimated amount is about 51 metric tonnes.

- Gas formation caused by anaerobic corrosion of steel and aluminium in SFL 3 from the waste packages is estimated to be about 19 830 m^3(STP)/year initially. The contribution from aluminium corrosion is 19 500 m^3(STP)/year, but will only last for the first three years after corrosion is initiated. At repository depth (500 m) the initial annual gas formation due to corrosion of steel and aluminium is about 400 m^3. After the aluminium has been consumed the annual gas formation is reduced to about 5 m^3.

- The gas formation rates in SFL 4 and SFL 5 are determined by the anaerobic corrosion of steel and are estimated to be about 1200 and 400 m^3(STP)/year initially.

- Gas formation due to radiolytic decomposition of water and microbial degradation was found to be of minor importance in all repository parts compared to gas formation by anaerobic corrosion.

- It has been established that waste containing organic material will be concentrated to SFL 3 and that the cellulose content will be low, about 0.5% of the total weight of the concrete in the waste packages. Cellulose can potentially generate complexing agents. Other substances capable of that are cyanides. However, the content of cyanide in the SFL 3 waste will be even lower, about 0.04% of the total weight of concrete in the waste packages.

- Laboratory experiments and calculations performed indicates that the leaching of concrete in the repository is slow and that pH will remain above 12.5 during at least the first 10^6 years.

- Sorption in concrete is an important barrier function that works for most of the radionuclides in the waste. The value of solubility retention is less well established but demonstrated to be effective for radionuclides of Ni (and Zr) in SFL 5.

- Solubilities of for example Ni in concrete cannot be calculated from existing thermodynamic databases, because there is a lack of constants relevant for high pH conditions. It is necessary to rely on direct measurements of solubilities.

- A horizontal distance of 1 km between SFL 3-5 and SFL 2 should prevent heat from SFL 2 to reach SFL 3-5 and it will also constitute a barrier to mass-transport between the two repository areas. However, it should be noted that certain chemical components from the SFL 3-5 repository such as concrete components, organic substances and gaseous compounds are not welcome in the canister positions. Therefore it seems worthwhile to create an additional barrier at a particular site through a favourable positioning of the repository parts regarding hydrology etc.

- So far no obvious disadvantages in the proposed design according to PLAN 93 has been identified. However, adding a barrier into SFL 4 which increases the retention and decreases the radionuclide releases from this repository part, may be worthwhile to consider. Also the dimensions of the tunnels used for storage in SFL 4 should be reconsidered. They seem to be a bit impractical in the present outline. Another option that may be worthwhile to consider is to exchange the bentonite barrier in SFL 3 by for example a sand or gravel backfill. At least it deserves to be tested if the bentonite barrier constitutes any significant advantage compared to a sand-gravel backfill.

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Abbreviations

α-Lab	Materials test laboratory (at Studsvik)
BWR	Boiling-Water Reactor
CLAB	Central interim storage for spent fuel
EP	Encapsulation Plant
FA	Fuel element storage (at Studsvik)
FEP	Features Events and Processes
FOA	National research defence institute
HCL	Hot Cell Laboratory (at Studsvik)
ILW	Intermediate Level Waste
IRM	Intermediate Range Monitoring
LLW	Low Level Waste
Package 1)	Packaging with its radioactive contents
Packaging 1)	The assembly of components necessary to enclose the radioactive contents completely, e.g. drum or container
PRM	Power Range Monitors
PS	Process System
PWR	Pressurized-Water Reactor
R1	Research reactor (at KTH)
R2	Research reactor (at Studsvik)
SFR	Final repository for low- and intermediate level radioactive waste (at the Forsmark Nuclear Power Station)
SFR 1	Final repository for radioactive operational waste
SFR 3	Final repository for decommissioning waste
SFL	Deep repository for long-lived radioactive waste
SFL 2	Deep repository for encapsulated spent fuel
SFL 3-5	Deep repository for long-lived low and intermediate level waste
SRM	Source Range Monitoring
TIP	Travelling Incore Probe

1) IAEA Safety Series No 6 "Regulations for the Safe Transport of Radioactive Material, 1985 edition"

Appendix A

Scenario Vocabulary

Scenario Vocabulary

There is often a problem with many words and terms used in performance assessments that general consensus of their meaning is lacking. To facilitate the reading of this report a short description of how words and terms are defined within the prestudy are presented. The definitions are to a large extent based on the Joint SKI/SKB Scenario Development Project [*Andersson, 1989*].

Scenario

Scenarios are used in many fields to describe possible future evolutions. In performance assessments of a repository a scenario is defined by a set of external conditions which will influence processes in a Process System. The external conditions determine how to actually model and combine the processes in the Process System when evaluating the consequence of the scenario [*Andersson, 1989*].

Process System

The Process System is the organized assembly of all phenomena (FEPs) required for description of barrier performance and radionuclide behaviour in a repository and its environment, and that can be predicted with at least some degree of determinism from a given set of external condition. The Process System hence comprises the complete set of "deterministic" chemical and physical processes that might influence the release of contaminants from the repository to the biosphere [*Andersson, 1989*].

Scenario methodology

The methodology used to select which scenario should be considered in the safety assessment. The scenarios chosen should have a broad coverage of all possible evolutions of the repository system. The methodology consists of several steps:

- a systematic written and visual description of the Process System
- a documented list of external events that might influence a repository and the relevance of their occurrence
- a documented description of how expert judgment has been used in the scenario methodology process
- a list of chosen scenarios and the motivation for the decision.

Scenario analysis

The actual analysis of a chosen scenario. This includes a written description of the scenario, the conceptual and numerical models used and the results from the calculations. A scenario analysis should also involve a uncertainty analysis.

Reference Scenario

A Reference Scenario is a scenario chosen for comparison reasons. It should not be seen as the most probable scenario. A Reference Scenario is often a simplified scenario which is rather easy to define and hence to perform calculations on.

Reference Case

In the quantitative analysis of the Reference Scenario a Reference Case is selected for comparison reasons. The Reference Case does not necessarily include all processes and mechanisms described in the Reference Scenario.

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Appendix B

Coprecipitation and adsorption on corrosion products

General remarks on coprecipitation processes

As a general case, trace substances are always coprecipitated with a precipitating major component if it fits into the crystal lattice of the major component [*Haissinsky, 1964*]. It is a prerequisite that the trace component crystals are isomorphous with the crystals of the major component.

According to [*Loberg, 1983*] a distinction must be made between polymorphic, isotypic and isomorphic crystals. Polymorphic crystals have identical chemical composition but different geometrical structure, e.g. aragonite and calcite, both which have the chemical composition $CaCO_3(s)$. Isotypic crystals have different chemical composition, but are similar in geometrical structure, e.g. $CaCO_3(s)$ and $CdCO_3(s)$. The condition for crystals to be isomorphic is that the atoms are able to replace each other in their respective lattices, i.e. to form solid solutions. This requires the crystals to be similar in geometrical structure, but in addition that the chemical bonds are of the same character, i.e. ionic or covalent bonds.

Coprecipitation may occur also when isomorphous crystals are not formed. Numerous types of coprecipitation processes are summarised by [*Beneš and Majer, 1980*]. According to the approximate Peneth-Fajans rule, a trace substance is adsorbed on a solid precipitate, or is coprecipitated with it, if the trace substance forms a low soluble compound with the counterion of the solid precipitate. Adsorption and coprecipitation are both favoured by a large surface area of the solid precipitate, and are favoured also if the surface has a surface charge of the opposite sign to that of the ionic trace substance.

Mechanistic description of coprecipitation and adsorption processes

In the following the term coprecipitation is used for systems where mixed crystals are formed when a major component is precipitated, carrying with it some trace substances which may be present at concentrations less than their solubility limits. This means that the trace substances enter into a solid state despite the fact that the thermodynamical solubility limit, as a pure substance, may not be exceeded. Many radionuclides present in ground water at trace levels may, therefore, be expected to be carried by precipitating major components, despite a low concentration, well below the solubility concentration. If such processes occur in a nuclear waste repository in deep ground waters, a significant retention may be expected for many of the radioelements present at trace levels. The definition of adsorption used in this study is a process where a dissolved trace substance interacts with the surface of an existing solid phase (e.g. a corrosion product) following some equilibrium condition which varies with the composition of the aqueous solution, e.g. the K_d-concept, Langmuir isotherm etc.

According to [*Haissinsky, 1964*], a dissolved substance tends to be distributed between the solution and the solid phases, so that its chemical potential is equal in the solution and in the solid phases. This property was frequently used in the past as a laboratory separation method for radioelements. The underlying mechanisms have been a subject of much debate, although some agreement on the mechanisms seems to have been reached today.

Theories are available to explain that the solubility of a trace substance is greatly reduced when appearing as a minor component in a solid solution [*Stumm, 1992*]. The reason for this is that the activity coefficient of the solid phase in a solid solution is not equal to 1. When comparing the measured solubilities with the solubilities calculated for the pure phases, it is often found that the solubility of the major component is only slightly affected by the formation of solid solutions, whereas, the solubility of the trace substance is significantly reduced. A distribution coefficient, D, can be formulated for a solid solution between two components MY and NY:

$$D = K_{S0}^{MY} \cdot \gamma^{MY} / (K_{S0}^{NY} \cdot \gamma^{NY}) \qquad \text{(Eq. 1)}$$

where:
K_{S0}^{i} = solubility product of component *i*
γ^{i} = activity of the solid phase *i* in the solid solution

In an ideal solid solution the quotient $\gamma^{MY} / \gamma^{NY}$ is equal to 1. An estimate of the activity quotient can be obtained by comparing calculated values with experimental measurements of the distribution coefficient D. This can be formulated as:

$$D_{\text{ideal}} = D_{\text{experimental}} \cdot \gamma^{\text{trace component}} / \gamma^{\text{major component}} \qquad \text{(Eq. 2)}$$

Recalling that for the major component the activity coefficient is close to unity, we find that:

$$\gamma^{\text{trace component}} = D_{\text{ideal}} / D_{\text{experimental}} \qquad \text{(Eq. 3)}$$

The chemical equilibria in a two-component solid solution system can be described by the following equations:

$$a_M \cdot a_Y = K_{S0}^{MY} \cdot X^{MY} \cdot \gamma^{MY} \qquad \text{(Eq. 4)}$$

$$a_N \cdot a_Y = K_{S0}^{NY} \cdot X^{NY} \cdot \gamma^{NY} \qquad \text{(Eq. 5)}$$

where:
a_j = activity if ion *j* in solution
X^{i} = the mole fraction of component *i* in the solid solution

Further we may recognize that the left hand side of equation 4 is equal to an apparent solubility constant, $K_{S0}^{*MY} = a_M \cdot a_Y$. As a first approach, we may assume a close to ideal behaviour of the trace component in dilute solid solutions (this assumption may need to be revised), i.e. $\gamma^{MY} \approx 1$. From this we find:

$$K_{S0}^{*MY} = K_{S0}^{MY} \cdot X^{MY} \qquad \text{(Eq. 6)}$$

A law governing the equilibrium distribution between the aqueous and solid phases can be obtained as the quotient between equations 4 and 5 and by inserting into equation 1. This law, known as the Berthelot-Nernst law, applies to homogeneous solid solutions:

$$X^{MY} / X^{NY} = D \cdot a_M / a_N \qquad \text{(Eq. 7)}$$

However, not all coprecipitation systems form homogeneous solid solutions. If adsorption is an active part of the overall process, a heterogeneous solid phase is formed, at least temporarily, with a concentration gradient in the solid phase from the surface to the centre of the crystals. For such a system the surface equilibrium system can best be described by the Doerner-Hoskins logarithmic distribution relationship:

$$\log \frac{[M_{initial\ solution}]}{[M_{final\ solution}]} = D' \cdot \log \frac{[N_{initial\ solution}]}{[N_{final\ solution}]} \qquad \text{(Eq. 8)}$$

where:
D' = constant

More advanced methods to describe and represent coprecipitation systems have been described elsewhere, e.g. [*Lippmann, 1980*] and [*Kornicker, 1991*], but will not be further treated in this study.

Experimental investigations and natural analogue studies

Experimental investigations of coprecipitation processes are a necessary complement to the thermodynamic theories described previously to judge the potential retention of trace elements. Experimental investigations are required both to increase the knowledge of the fundamental mechanisms governing coprecipitation and to evaluate the magnitude of coprecipitation in a particular system.

Several difficulties in studies of coprecipitation processes can be identified, e.g.:
- kinetic effects,
- adsorption phenomena,
- recrystallisation phenomena, and
- variations in the chemical environment.

The kinetic effects are difficult to evaluate, some reactions may be very slow, so that true equilibrium is not reached even on geological time scales. However, also much shorter chemical changes may influence the results significantly. Consequently, the short time scales in experiments could give a wrong picture of the long term situation to be expected in the geological system of interest for nuclear waste storage.

The adsorption of trace elements on solid precipitates is a well known process. Adsorption is usually the first step in the coprecipitation process. The adsorption is favoured by precipitates with large surface area, a good example is the adsorption onto freshly precipitated iron oxyhydroxides frequently used also in commercial water cleaning systems. This type of transfer to solid form is not necessarily a true coprecipitation effect which will form a solid solution. Thus, ageing effects could be pronounced whereby in some cases the trace elements may be released into solution after a certain time.

Recrystallisation means a transformation of the crystal geometry into a form with a more stable configuration. As was discussed previously a rearrangement of the crystal structure of the major component may have the effect that the trace element no longer fits into the crystal lattice. As a result, a solid solution is no longer formed, and the trace element may be released into solution, or may be enriched at the surface of the mineral grains. This is an important type of ageing processes which must be regarded when evaluating coprecipitation processes observed during short-term experiments. An experimental study related to this has been presented by *[Thornber and Wildman, 1984]*.

It is quite obvious that laboratory conditions will never resemble those of the geochemical system in deep rock. The complexity of the natural mineral assemblies and groundwaters may be simulated, but the slow evolution of the conditions which may have occurred during thousands of years in the geochemical systems can hardly be expected to be treated reliably in the laboratory.

Studies of natural analogues could be valuable in this sense, where different processes can be studied in nature. The study of natural analogues is valuable since it offers a unique possibility to observe and evaluate the importance of long term effects, thereby approaching the crucial effects of kinetics in many geochemical systems. However, the high complexity of most natural systems introduces large difficulties in the interpretation, not the least when considering the possibilities to distinguish between effects of different processes. Nevertheless, some excellent studies have recently been published on the natural analogue site at Poços de Caldas, e.g. [*Bruno et al., 1991*], in which systematic approaches have been taken to evaluate different phenomena. Results indicate that concentrations of nickel, zinc and manganese may be controlled by coprecipitation with ferric oxyhydroxides.

Coprecipitation has been observed when manganese is oxidised to Mn(IV) in the presence of Co^{2+}, Zn^{2+}, Cu^{2+} and Ni^{2+}. The systems have been studied for different conditions and have been evaluated in different ways. It has been found that coordination chemical properties of the metal ions involved in the reactions are helpful tools to explain and predict the results [*Hem and Lind, 1991*].

Studies of trace metal concentrations in sulphidic waters show that dissolved metal concentrations are commonly not in equilibrium with pure metal sulphides [*Arakaki and Morse, 1993*]. The deviation has been interpreted as a result of adsorption or coprecipitation.

Aspects on the long-term properties of coprecipitation systems

A critical question that need to be addressed is the long-term stability of the coprecipitates/solid solutions formed initially. In general, coprecipitation processes which only results in a temporary capturing of radioelements are of little interest for a performance assessment study. The formation of true solid solutions may on the other hand completely immobilise the trace elements for as long as the solid phase of the major component exists. The coprecipitation of trace elements with ferric hydroxide is an example where the long-term properties may change due to recrystallisation of the carrier substance. Trace elements which are not compatible with the recrystallised phase, e.g. crystalline goethite, would then be expected to be released into solution or precipitate as separate crystals. If on the other hand, the trace element can form a solid solution also with the recrystallised ferric hydroxide, a more stable solid product is formed with long-term properties likely to be improved.

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Appendix C

Experiments at CTH

Experiments at CTH

The chemical condition in a concrete based repository is dependent on the long-term stability of the concrete. The solubility and the sorption properties of some of the radionuclides is dependent on pH and ionic strength in the pore water. Also, the long term performance in terms of hydraulic resistance of such a barrier is dependent on the leach rate of the cement paste in the concrete.

Experimental activities

Sorption studies

General

In the process of producing the sorption database for this prestudy, it was in some cases necessary to verify literature values by performing experiments. In other cases, where values based on experiments were missing and the values assigned were from chemical analogues, experiments were needed.

Experimental

Concrete from the silo and porous cement paste from SFR was crushed and sieved in a nitrogen filled glove box. A fraction with a grain size of 0.065-0.355 mm was collected and used for the sorption experiments. As the aqueous phase, a synthetic cement porewater with a pH of 13.5 was used. These experimental conditions represent the sorption in fresh concrete or cement paste.

The samples were stored in the glove box and gently agitated by rotating slowly. The measurements of the sorption were performed after one week, one month, three months and one year. For phase separation, a high speed centrifuge with a centrifugal field of 13 000 G was used.

Results

The sorption of nickel on concrete and cement paste has been studied previously [*Allard and Andersson, 1987*]. The K_d-value reported was 1 m^3/kg. However, new sorption experiments at lower nickel concentrations yield lower values. According to these results we recommend the value $K_d = 0.1$ m^3/kg in Table 7.2. A possible explanation to the higher values in earlier experiments may me supersaturation of nickel because it was difficult to control the concentration of nickel.

The sorption of radium in Table 7.2 is selected from Bradbury et al. [*Bradbury and Sarott, 1994*]. The authors claim however that there are some uncertainties in the original work on which their value is based. In order to verify their selection, some simple sorption studies were performed on Ra. The results support the choice of a K_d-value of 0.5 m^3/kg.

In Table 7.2, a K_d-value of 0.1 m^3/kg is selected due to its similarity to nickel. This choice has been supported by experiments with ^{60}Co. The measured sorption on concrete and cement paste ranges between 0.2 to 0.4 m^3/kg.

Experiments with ^{234}Th indicate the difficulties of determining "true" sorption of tetra valent actinides due to their low solubility. Although it was possible to work with a very low concentration of Th when using ^{234}Th ($t_{1/2}$ =24 d), no reliable results were obtained. The problems are probably of different nature. First, the actinides readily form colloids in supersaturated solutions and those are difficult to separate from the aqueous phase. Secondly, it is difficult to distinguish "true" sorption and precipitation on the solid surface. But to conclude, the sorption is very strong and for the moment there is no reason to revise the sorption data for the actinides.

Leaching experiments

In order to study the leach rate of cement, an investigation of static leaching of cement paste is being performed. The influence of the leaching water composition (saline and non-saline) on the leaching process is being studied. In this initial phase, cement paste contacted leach water and not the solid phase is being analysed.

Experimental

Hydrated Ordinary Portland Cement paste (Degerhamn anläggningscement) was crushed and sieved and a grain size of 0.063 to 0.125 mm was collected. The cement was moulded in 1986 and stored in water until 1993. The cement paste was considered fully hydrated since it had been stored such a long time in an aqueous phase.

Six polypropylene tubes were prepared with 5 g of the solid material and 20 ml of synthetic groundwater was added to each tube. In three of the tubes the added ground water was typical non-saline water, that is Allard Water (c.f. composition in table C1). In the other three, a saline water composed by Nagra and SKB (NASK, c.f. composition in table C1), was added. The material was crushed and the tubes are stored in a nitrogen filled glove box with a low content of carbon dioxide.

The water in each tube was partially replaced (typically 12 ml) by fresh water every week. The collected water was analysed for the content of sodium, potassium, calcium, magnesium, chloride, nitrate, sulphate, silicone and hydroxide.

Table C1 Composition of the synthetic ground water.

	Concentration (m/l)	
	Allard water	NASK water
Na^+	65	3220
K^+	3.9	80
Ca^{2+}	18	800
M^{2+}	4.3	10
Cl^-	70	6390
HCO_3^-	123	120
SO_4^{2-}	9.6	380
H_4SiO_4	12	
pH	8.0-8.2	7.7

Results

By studying the pH development as a function of the number of leaching steps, it is possible to evaluate the long-time performance of an engineered concrete barrier. As can be seen from figures C1 and C2, the period of a pH of about 12.5 lasts at least 45 leaching steps. This period corresponds to about a million years assuming a constant porosity of the solid throughout the leaching process (c.f. Table C2)

Table C2 Physical and experiential data and data assumed on the crushed cement paste

Cement paste	Ordinary Portland Cement (Degerhamn Anläggningscement)
Hydration time	7 years
Water cement ratio w/c	0.5
Bulk density	2770 kg/m^3
Amount of crushed cement	5.0 g
Temperature	22°C
Grain size	0.065-0.125 mm
Porosity	30 %
Total pore volume in 5g material	0.54 ml
Volume of water replaced in each step	12 ml
Number of pore volumes replaced in each step	22
Assumed time for one pore volume replacement in repository conditions	1000 years

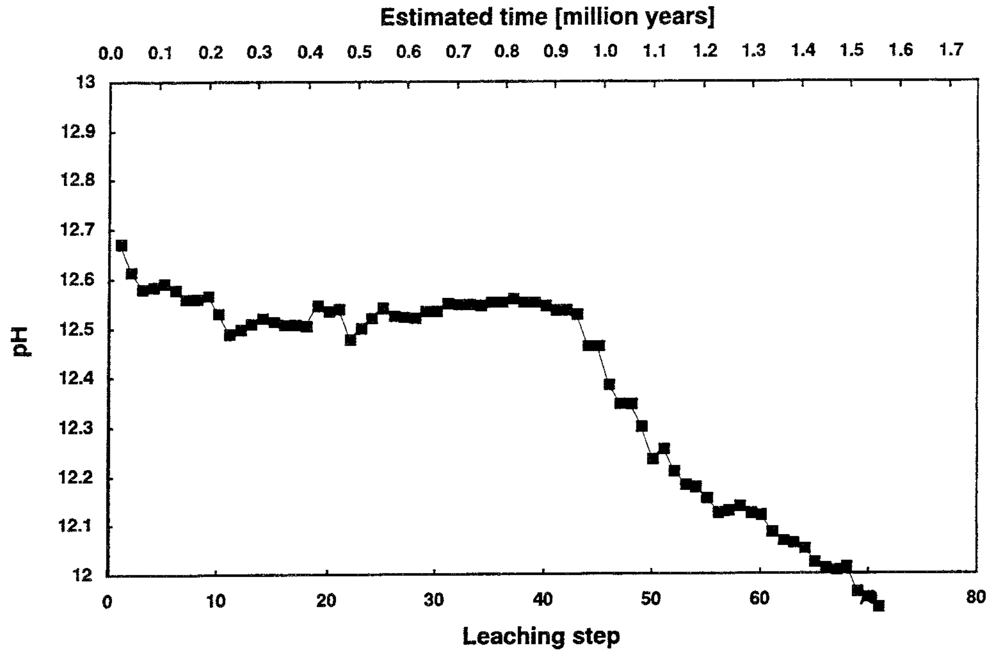

Figure C1 *pH development of leach water in the NASK experiments (mean value of three experiments).*

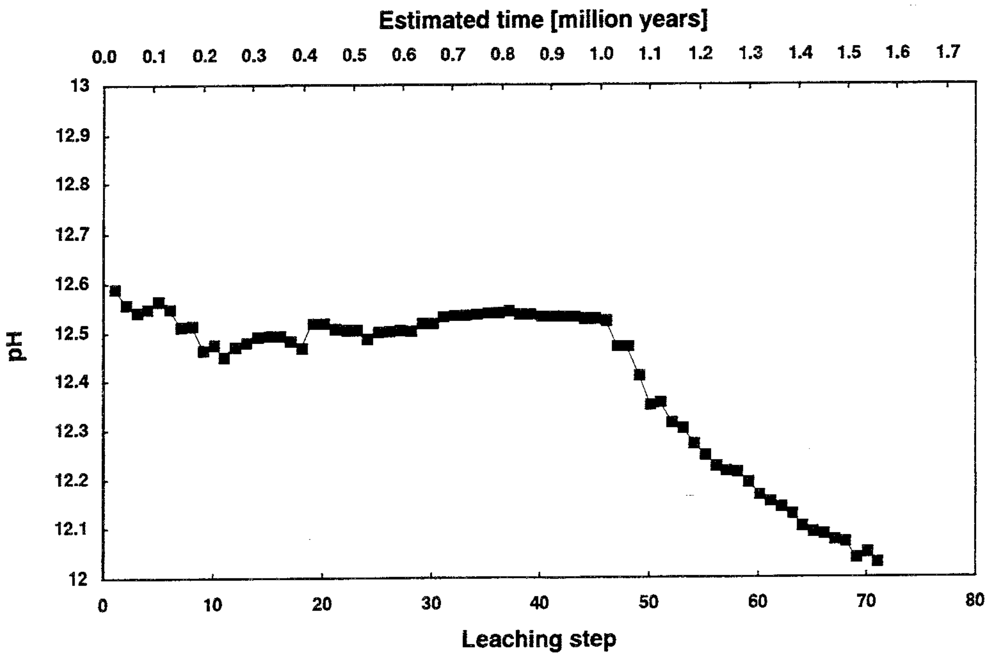

Figure C2 *pH development of leach water in the Allard experiments (mean value of three experiments).*

The assumption of the material having a constant porosity of 30% is supported by a mass balance over 70 steps in the Allard experiment. Calculations show that only 10% of the solid phase is dissolved. This supports the assumption of a constant porosity. However, this calculation is an overall mass balance and does not account for any dissolution/precipitation. The dissolution of the initial material is larger but is compensated by the formation of secondary phases

The period of pH about 12.5 to 12.6 indicate the dissolution of portlandite ($Ca(OH)_2$) since the mineral buffers a solution at that pH-value. That period is followed by the incongruent dissolution of the calcium silicate hydrate C-S-H gel. During this period, the Ca/Si ratio in the solid is decreasing from the initially 1.7 to 0.8 by depletion of Ca. After that a period of pH about 11 is expected, buffered by the congruent dissolution of the hydrated components.

Conclusion

A conclusion from the results is that only the period of fresh and leached concrete ($12 < pH < 13.5$) has to be considered while selecting solubilities and sorption properties of the radionuclides and toxic elements present in the repository. The period of degraded concrete seems to be irrelevant. This is true for non-saline and saline conditions.

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