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Fuel and canister process report for the safety assessment SR-Can

Svensk Kärnbränslehantering AB

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Preface

This document compiles information on processes in the fuel and in the canister relevant for long-term safety of a KBS-3 repository. It supports the safety assessment SR-Can, which is a preparatory step for a safety assessment that will support the licence application for a final repository in Sweden.

The report was edited by Lars Werme, SKB.

The following persons have had the main responsibilities for specific subject areas: Lars Werme, SKB (canister and part of fuel processes), Kastriot Spahiu (part of fuel processes), Patrik Sellin (part of fuel processes). The specific parts of their contributions are listed in Section 1.3 in the report.

The report has been externally reviewed by David Shoesmith, the University of Western Ontario, Peter Dillström, Inspecta Technology AB, and Fraser King, Integrity Corrosion Consulting Ltd.

Stockholm, October 2006

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Contents

1 Introduction

1.1 Role of this process report in the SR-Can assessment

1.1.1 Overall methodology

This report documents fuel and canister processes identified as relevant to the long-term safety of a KBS-3 repository. It forms an important part of the reporting of the safety assessment SR-Can. The detailed assessment methodology, including the role of the process report in the assessment, is described in the SR-Can Main report /SKB 2006a/. The following excerpts describe the methodology and clarify the role of this process report in the assessment.

The repository system broadly defined as the deposited spent nuclear fuel, the engineered barriers surrounding it, the host rock, and the biosphere in the proximity of the repository will evolve over time. Future states of the system will depend on:

- the initial state of the system,
- a number of radiation related, thermal, hydraulic, mechanical, chemical and biological processes acting within the repository system over time, and
- external influences acting on the system.

A methodology in ten steps has been developed for SR-Can as summarised in Figure 1-1 and described below.

1. Identification of factors to consider (FEP processing).

   This step consists of identifying all the factors that need to be included in the analysis. Experience from earlier safety assessments and KBS-3 specific and international databases of relevant features, events and processes (FEPs) influencing long-term safety are utilised. An SKB FEP database is developed where the great majority of FEPs are classified as being either initial state FEPs, internal processes or external FEPs. Remaining FEPs are either related to assessment methodology in general or deemed irrelevant for the KBS-3 concept. Based on the results of the FEP processing, an SR-Can FEP catalogue, containing FEPs to be handled in SR-Can, has been established. This step is documented in the SR-Can FEP report /SKB 2006c/.

2. Description of the initial state.

   The initial state of the system is described based on the design specifications of the KBS-3 repository, a descriptive model of the repository site and a site-specific layout of the repository. The initial state of the fuel and the engineered components is that immediately after deposition, as described in the SR-Can Initial state report /SKB 2006d/. The initial state of the geosphere and the biosphere is that of the natural system prior to excavation, as described in the site descriptive models /SKB 2005, 2006j/. The repository layouts adapted to the sites are provided in underground design reports for each site /Brantberger et al. 2006, Janson et al. 2006/.

3. Description of external conditions.

   Factors related to external conditions are handled in the categories "climate related issues", "large-scale geological processes and effects" and "future human actions". The handling of climate related issues is described in the SR-Can Climate report /SKB 2006e/, whereas the few external, large-scale geosphere processes are addressed in the Geosphere process report. The treatment of future human actions in SR-Can is described in the SR-Can FHA report /SKB 2006f/.

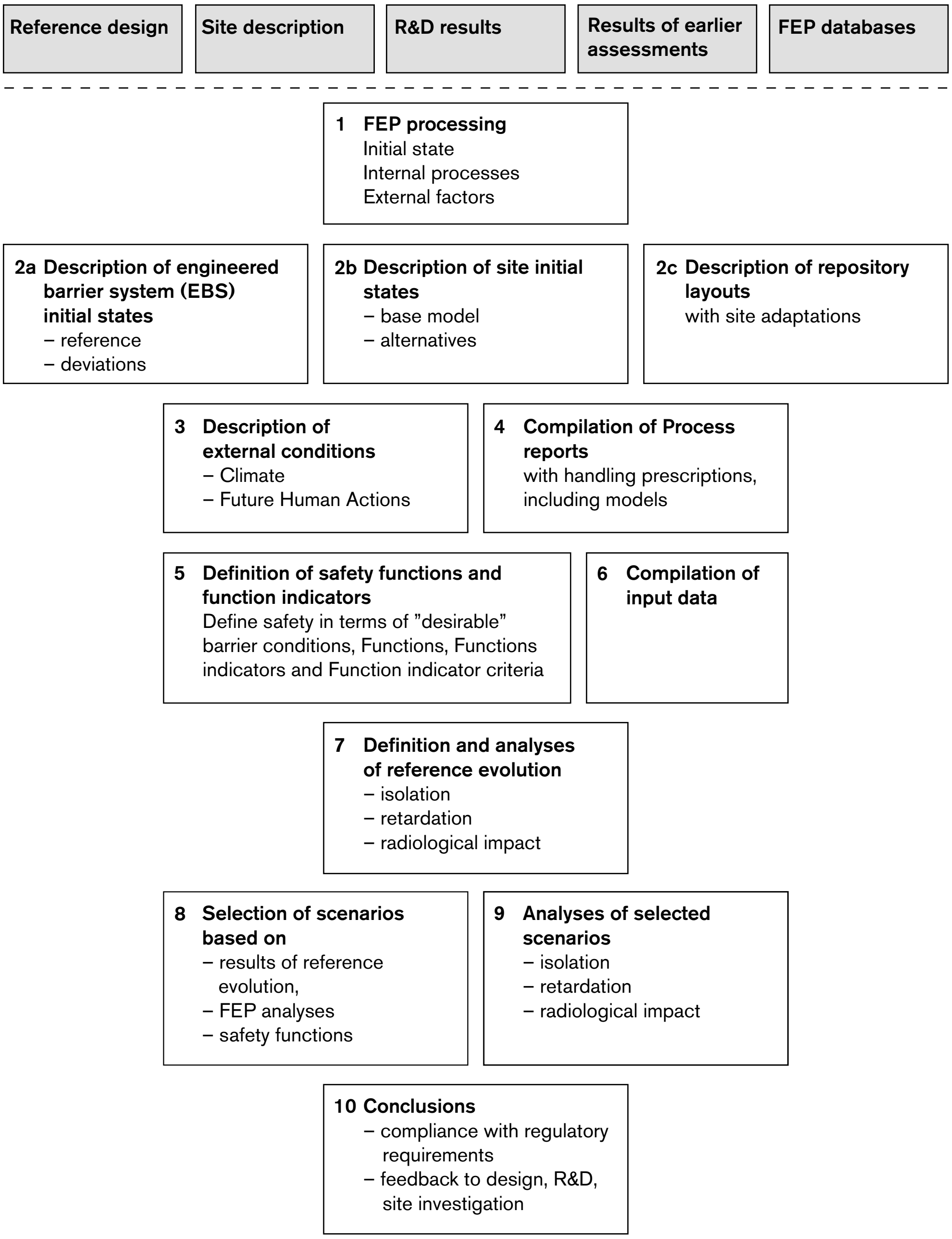

Figure 1-1. *An outline of the ten main steps of the SR-Can safety assessment. The boxes at the top above the dashed line are inputs to the assessment.*

4. Description of processes.

   The identification of relevant processes is based on earlier assessments and FEP screening. All identified processes within the system boundary relevant to the long-term evolution of the system are described in dedicated Process reports, i.e. this one for the fuel and canister, for the buffer and backfill /SKB 2006g/, for the geosphere /SKB 2006h/, and for the biosphere /SKB 2006i/. Also short-term geosphere processes/alterations due to repository excavation are included. For each process, its general characteristics, the time frame in which it is important, the other processes to which it is coupled and how the process is handled in the safety assessment are documented.

5. Definition of safety functions, function indicators and function indicator criteria.

   This step consists of an account of the safety functions of the system and of how they can be evaluated by means of a set of function indicators that are, in principle, measurable or calculable properties of the system. Criteria for the safety function indicators are provided. The Process reports are important references for this step. A FEP chart is developed, showing how FEPs are related to the function indicators.

6. Input data selection.

   Data to be used in the quantification of repository evolution and in dose calculations are selected using a structured procedure. The process of selection and the data adopted are reported in a dedicated Data report /SKB 2006b/. Also, a template for discussion of input data uncertainties has been developed and applied.

7. Definition and analysis of reference evolution.

   A reference evolution, providing a description of a plausible evolution of the repository system, is defined and analysed. The isolation potential of the system over time is analysed in a first step, yielding a description of the general system evolution and an evaluation of the safety function indicators. If the evolution indicates breaching of isolation, the retardation potential of the repository and its environs is analysed and dose consequences are calculated for the long-term conditions identified in the first step. Also, some canister failure modes not resulting from the reference evolution are analysed in order to further elucidate the retarding properties of the system. Each process is handled in accordance with the plans outlined in the process reports.

8. Selection of scenarios.

   A set of scenarios for the assessment is selected. A comprehensive main scenario is defined in accordance with SKI's regulations SKIFS 2002:1. The main scenario is closely related to the reference evolution analysed in step 7. The selection of additional scenarios is focused on the safety functions of the repository and the safety function indicators defined in step 4 form an important basis for the selection. For each safety function, an assessment is made as to whether any reasonable situation where it is not maintained can be identified. If this is the case, the corresponding scenario is included in the risk evaluation for the repository, with the overall risk determined by a summation over such scenarios. The set of selected scenarios also includes, e.g. scenarios explicitly mentioned in applicable regulations, such as human intrusion scenarios, and scenarios and variants to explore design issues and the roles of various components in the repository.

9. Analysis of scenarios.

   The main scenario is analysed essentially by referring to the reference evolution in step 7. An important result is a calculated risk contribution from the main scenario. The additional scenarios are analysed by focusing on the factors potentially leading to situations in which the safety function in question is not maintained. In most cases, these analyses are carried out by comparison with the evolution for the main scenario, meaning that they only encompass aspects of repository evolution for which the scenario in question differs from the main scenario. For these scenarios, as for the main scenario, a risk contribution is estimated.

10. Conclusions.

    This step includes integration of the results from the various scenario analyses, development of conclusions regarding safety in relation to acceptance criteria and feedback concerning design, continued site investigations, and R&D programme.

This Fuel and Canister process report is one of the process reports mentioned in step 4 above. The purpose of the process reports is to document the scientific knowledge of the processes to a level required for an adequate treatment of the processes in the safety assessment. The documentation is not exhaustive from a scientific point of view, since such a treatment is neither necessary for the purposes of the safety assessment nor possible within the scope of an assessment. However, it must be sufficiently detailed to motivate, by arguments founded on scientific understanding, the treatment of each process in the safety assessment. The purpose is further to determine how to handle each process in the safety assessment at an appropriate degree of detail, and to demonstrate how uncertainties are taken care of, given the suggested handling. The handlings established in this report are used in the analysis of the reference evolution, step 7 and in the analyses of scenarios, step 9.

1.1.2 Identification of processes

The process documentation in the SR 97 version of the Process report /SKB 1999/ is a starting point for this SR-Can version. As a result of the FEP processing (step 1 above), the list of relevant processes has been reviewed and slightly extended, see the SR-Can FEP report /SKB 2006c/ for details. Furthermore, the SR 97 process report covering fuel, canister, buffer/backfill and geosphere processes has been replaced by several reports of which the present one covers fuel and canister processes. A complete list of processes described in the SR-Can process reports can be found in the abovementioned FEP report.

1.1.3 Relation to specific sites

The SR-Can assessment builds on site-specific data and site-descriptive models of the Forsmark area /SKB 2005/ and the Laxemar subarea /SKB 2006j/, which currently are being investigated as candidate sites for a final repository for spent nuclear fuel in Sweden, see further the SR-Can Main report /SKB 2006a/. Geosphere data for SR-Can are obtained from the site-descriptive model versions 1.2, which are based on the initial site investigation stage. The process descriptions are of a sufficiently general nature to be applicable to both these sites. However, the result of the quantitative evaluations of the processes in the different scenarios analysed in SR-Can will, in many cases, be dependent on site-specific data. These data are not given here, but in dedicated model studies. In addition, the most essential data for the safety assessment are thoroughly evaluated in the SR-Can Data report /SKB 2006b/, step 6 above.

1.1.4 Intended audience for this report

This report is written by, and for, experts in the relevant scientific fields. It should though be possible for a generalist in the area of long-term safety assessments of geologic nuclear waste repositories to comprehend the contents of the report. The report is an important part of the documentation of the SR-Can project and an essential reference within the project, providing a scientifically motivated plan for the handling of geosphere processes. It is, furthermore, foreseen that the report will be essential for reviewers scrutinising the handling of geosphere issues in the SR-Can assessment.

1.2 Structure for process descriptions

All identified processes are documented using a template, where many of the headings are the same as those used in the SR 97 report. These are described below.

Overview/general description

Under this heading, a general description of the knowledge regarding the process is given. For most processes, a basis for this is the contents of the SR 97 Process report /SKB 1999/, but reviewed and updated as necessary.

Dependencies between process and fuel and canister variables

For each system component, in this case the fuel and canister systems, a set of physical variables that defines the state of the system is specified. For each process, a table is presented under this heading with documentation of how the process is influenced by the specified set of physical variables and how the process influences the variables. In addition, the handling of each influence in SR-Can is indicated in the table.

Boundary conditions

The boundary conditions for each process are discussed. These refer to the boundaries of the fuel and canister systems, respectively. The processes for which boundary conditions need to be described are, in general, related to transport of material or energy across the boundaries.

Model studies/experimental studies

Model and experimental studies of the process are summarised. This documentation constitutes the major source of information for many of the processes.

Time perspective

The time scale or time scales on which the process occurs are documented, if such timescales can be defined.

Natural analogues/observations in nature

If relevant, natural analogues and/or observations in nature that contribute to the present understanding of the process are documented under this heading.

Handling in the safety assessment SR-Can

Under this heading, the handling in the safety assessment SR-Can is described. Typically, the process is either

- neglected on the basis of the information under the previous headings, or
- included by means of modelling.

The following aspects need to be covered, although no prescribed format for the documentation is given:

Boundary conditions: How are the boundary conditions handled? For example, are spatially and temporally varying chemical and hydraulic conditions considered?

Influences and couplings to other processes: The handling of the documented influences will be discussed as will couplings to other processes within the system component.

As a result of the information under this subheading, a mapping of all processes to method of treatment and, in relevant cases, applicable models will be produced for all processes.

Handling of uncertainties in SR-Can

Given the adopted handling in the safety assessment SR-Can as described above, the handling of different types of uncertainties associated with the process will be summarised.

Uncertainties in mechanistic understanding: The uncertainty in the general understanding of the process is discussed based on the available documentation and with the aim of addressing whether the basic scientific mechanisms governing the process are understood to the level necessary for the suggested handling.

Model simplification uncertainties: In most cases, the quantitative representation of a process will contain simplifications. These may be a significant source of uncertainty in the description of the system evolution. These uncertainties are discussed and approaches to addressing them are identified including alternative models or alternative approaches to simplification of a particular conceptual model.

Input data and data uncertainties: The set of input data necessary to quantify the process for the suggested handling is documented. The further treatment of important input data and input data uncertainties is described in a separate report, the SR-Can Data report /SKB 2006b/, to which reference is made when relevant.

References

A list of references used in the process documentation is given at the end of the report.

1.3 Documentation of participating experts and of decisions made

The experts involved in assembling the basic information about the processes are listed in Table 1-1.

In the final version of the SR-Can process report, each process description will contain a documentation of which expert(s) assembled the basic information on the process, which expert(s) were involved in the decision regarding treatment in the safety assessment and the dates for the final revision of the text and for the final decision on handling. All these experts will be included in the forthcoming SR-Can expert database, see further the SR-Can main report /SKB 2006a/.

Table 1-1. Experts responsible for the process documentations.

Fuel processes	Expert author, affiliation
Radioactive decay	Lars Werme, SKB
Radiation attenuation/heat generation	Lars Werme, SKB
Induced fission (criticality)	Lars Werme, SKB
Heat transport	Lars Werme, SKB
Water transport in the canister cavity, boiling/condensation	Lars Werme, SKB
Mechanical cladding failure	Lars Werme, SKB
Structural evolution of the fuel matrix	Lars Werme, SKB
Advection and diffusion	Lars Werme, SKB
Residual gas radiolysis/acid formation	Lars Werme, SKB
Water radiolysis	Kastriot Spahiu, SKB
Metal corrosion	Lars Werme, SKB
Fuel dissolution	Kastriot Spahiu, SKB
Dissolution of the gap inventory	Kastriot Spahiu, SKB
Speciation of radionuclides, colloid formation	Kastriot Spahiu, SKB
Helium production	Lars Werme, SKB

Canister processes	**Expert author, affiliation**
Radiation attenuation/heat generation	Lars Werme, SKB
Heat transport	Lars Werme, SKB
Deformation of the cast iron insert	Lars Werme, SKB
Deformation of the copper canister from external pressure	Lars Werme, SKB
Thermal expansion (both cast iron insert and copper canister)	Lars Werme, SKB
Deformation from internal corrosion products	Lars Werme, SKB
Radiation effects	Lars Werme, SKB
Corrosion of the cast iron insert	Lars Werme, SKB
Galvanic corrosion	Lars Werme, SKB
Stress corrosion cracking of the cast iron insert	Lars Werme, SKB
Corrosion of the copper canister	Lars Werme, SKB
Stress corrosion cracking of the copper canister	Lars Werme, SKB
Earth currents – Stray current corrosion	Lars Werme, SKB
Deposition of salts on the canister surface	Lars Werme, SKB
Radionuclide transport	Patrik Sellin, SKB

1.4 Initial state of the fuel in SR-Can

The following is an overview description of the initial state of the fuel, i.e. its state at the time of deposition. Additional information is given in the SR-Can Initial State Report /SKB 2006d/.

1.4.1 General

This repository sub-system comprises the spent fuel and the cavity in the canister. The total quantity of spent fuel obtained from the Swedish nuclear reactors will depend on operating time, energy output and fuel burn-up. At the beginning of 2003 approximately 5,700 tonnes of spent fuel have been generated /SKB 2003/. With an operating time of 40 years for all reactors, except for Barsebäck 1 which was taken out of operation during 1999, the total quantity of spent fuel can be estimated at 9,500 tonnes /SKB 2003/.

1.4.2 Fuel types

Several types of fuel will be emplaced in the repository. For the reference case with 40 years of reactor operation, the fuel quantity from boiling water reactors, BWR fuel, is estimated at 7,000 tonnes, while the quantity from pressurized water reactors, PWR fuel, is estimated at about 2,300 tonnes /SKB 2003/. In addition, 23 tonnes of mixed-oxide fuel (MOX) fuel of German origin from BWR and PWR reactors and 20 tonnes of fuel from the decommissioned heavy water reactor in Ågesta will be disposed of. The total amount of spent fuel and the total number of fuel elements are given in Table 1-2.

Fuel burn-ups are expected to vary from about 15 MWd/kgU up to 60 MWd/kg /SKB 2004a/. The burn-up distribution in Clab in June 2005 is given in Figure 1-2.

Differences between different fuel types are important for criticality assessments.

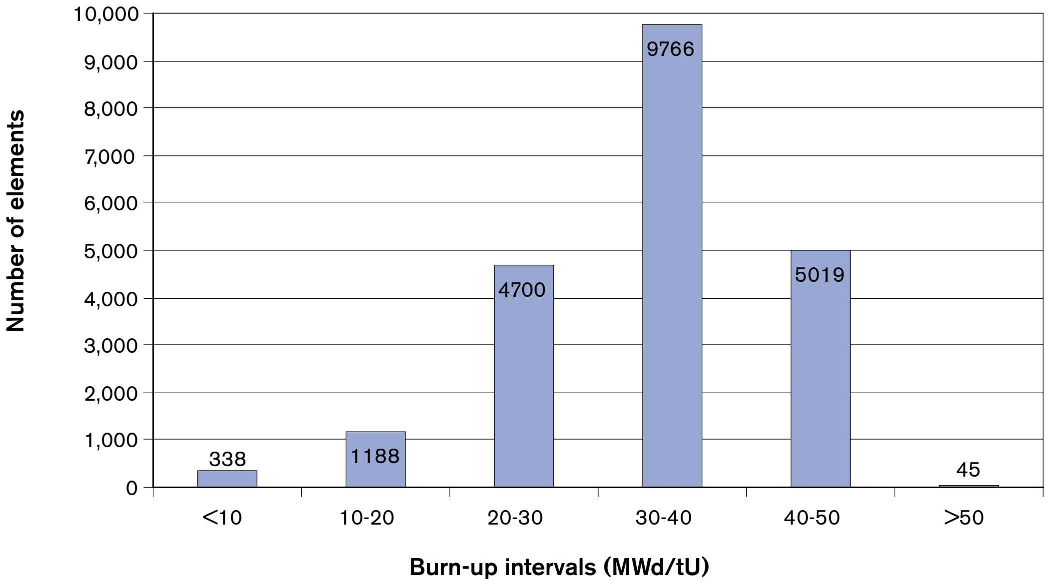

Figure 1-2. *Burn-up distribution in Clab (30 June 2005).*

Table 1-2. Amount of spent fuel and number of fuel elements.

Fuel type	Scenario with 40 years of reactor operation /Plan 2005/	
	Spent fuel (tonnes of U)	Number of fuel elements/units
BWR	7,038	38,740
PWR	2,310	5,000
MOX, Ågesta, Studsvik	23+20+2.4	640

To allow for future changes in the Swedish nuclear programme, the safety assessment assumes a total of 6,000 canister corresponding to 12,000 tonnes of fuel.

1.4.3 Structure of the fuel assemblies

Nuclear fuel, here exemplified by BWR fuel (Svea 96), consists of cylindrical pellets of uranium dioxide. The pellets are 11 mm high and have a diameter of 8 mm. They are stacked in approximately 4-metre-long cladding tubes of Zircaloy, a durable zirconium alloy. The tubes are sealed with welds and bundled together into fuel assemblies. Each assembly contains 96 cladding tubes. The fuel assembly also contains components of the nickel alloys Inconel and Incoloy, and of stainless steel. Pellets in a cladding tube and a fuel assembly are shown in Figure 1-3.

Aspects of importance in the safety assessment, for example geometrical aspects of the fuel cladding tubes, are as a rule handled sufficiently pessimistically in analyses of radionuclide transport that differences between different fuel types are irrelevant.

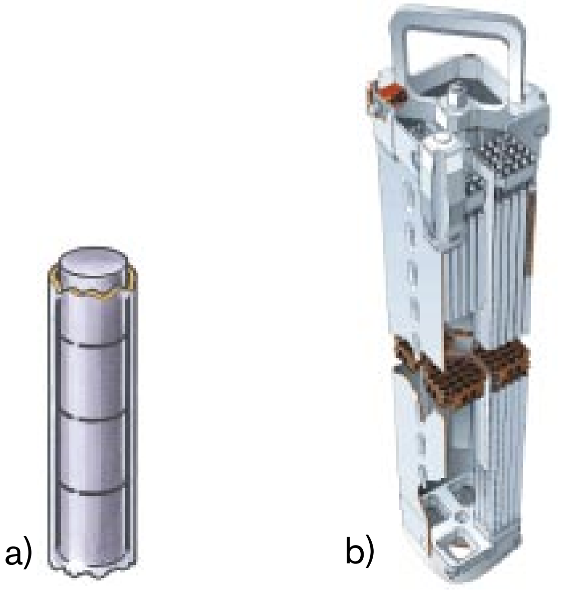

***Figure 1-3.** a. Cylindrical fuel pellets in cladding tubes of Zircaloy. The pellets have a diameter of approximately one centimetre. b. Fuel assembly of type Svea 96. The assembly consists of 96 fuel tubes and has a height of approximately 4 metres.*

1.4.4 Description of fuel structure and radionuclide distribution in the structure

Fuel

The nuclear fuel consists of very nearly stoichiometric uranium dioxide in the form of cylindrical pellets approximately 1 cm in diameter and 1 cm in length. The grain size is normally in the range of a few microns to about ten microns. The uranium in the fuel has been enriched in the isotope U-235 from the naturally occurring concentration of 0.7 percent up to 3.6 percent in BWR fuel (4.2 percent in gadolinium-bearing fuel) and 4.2 percent in PWR fuel.

The fuel typically develops a power of 15 to 25 kW per metre of fuel rod during operation, which corresponds to a centre temperature in the fuel pellet of 800 to 1,200°C. After use in the reactor, the fuel contains 3–5 percent fission products, depending on the burn-up, and approximately 1 percent higher actinides formed by neutron capture and radioactive decay. The average burn-up for Swedish nuclear fuel is currently (2003) 33 MWd/kg U, but the average burn-up has gradually increased and will be in the range 40–50 MWd/kg U in newer fuel.

The majority of the fission products and the higher actinides are present as a solid solution in the uranium dioxide matrix. Besides the chemical alteration of the uranium dioxide this entails, the fuel has also been altered physically during operation in the reactor.

Enrichment on pellet surface

In the case of fuel that has been irradiated at a relatively low temperature, the radial variation in grain size and porosity is small, with moderate grain growth in the centre of the pellet. An exception to this rule is the sharp increase in porosity in the pellet rim. To a depth of a few microns on the rim of the pellet, the porosity is several times higher than inside the fuel. This rim zone also deviates in microstructure from the rest of the pellet in that the original grains have been subdivided into many smaller grains. The reason for this lies in local variations in the fission rate across the diameter of the fuel.

Self-shielding in the fuel leads to a radial variation in the frequency of U-235 fissions, but more importantly, increased formation and fission of plutonium isotopes, in the rim of the fuel pellet. The result is increased burn-up and alpha activity at the periphery of the fuel pellets. The increased burn-up is also accompanied by a higher content of fission products. For an average burn-up of about 40 MWd/kg U, this surface layer can have a burn-up up to twice the average as well as more than four times the average alpha activity /Forsyth 1995/.

Radionuclides in the fuel-clad gap

During irradiation in the reactor, a fraction of the radionuclide inventory will have segregated either to the gap between the fuel and the cladding or to the grain boundaries. Of these radionuclides, the behaviour of the fission gases is best known. A number of studies of fission gas releases have been published over the years. The behaviour of other potentially segregated radionuclides is far less well-known. This is of concern because these radionuclides will be released more rapidly than the radionuclides that are embedded in the fuel's UO_2 matrix. The fraction of the radionuclides that is incompatible with the UO_2 matrix and, thus present in the fuel/cladding gap, is generally considered to be comparable to the fission gas release to the fuel/cladding gap as measured in gas release testing of fuel rods /Johnson and Tait 1997, Johnson and McGinnes 2002/. Since there are far more data available on fission gas release than on the release from the matrix of other radionuclides, it can be of interest to discuss the fission gas release even though the fission gases are of little concern for a long-term safety assessment. Fission gas release is more strongly correlated to the linear heat rating than to the burn-up of the fuel /Kamikura 1992/. Operating conditions ensure that the linear heat rating is kept relatively low and the fission gas release is minimized. Up to a burn-up of about 40 MWd/kgU, the fission gas releases are typically less than one percent /see e.g. Werme et al. 2004, Johnson et al. 2004/. About 75 percent of the Swedish fuel inventory has a burnup of less than 40 MWd/kgU and the number of fuel elements with an average burnup exceeding 50 MWd/kgU is very small, as of June 2005 only 45 fuel elements had a burnup exceeding 50 MWd/kgU. For the interval 40 to 50 MWd/kgU, the fission gas release is generally below 1.5% for PWR while for BWR it can go as high as 5% with an average in the range of 2.5%. There are very little data available for fission gas release from BWR fuel with higher burnup than 50 MWd/kgU. For PWR fuel the fission gas release for fuel in the burnup interval 50 to 60 MWd/kgU appears to several percent, pessimistically up to 5% /Werme et al. 2004, Johnson et al. 2004/.

There are relatively few systematic studies of the release of segregated radionuclides other than fission gases in fuel. By far the most comprehensive study was published by /Stroes-Gascoyne 1996/. In that publication, the rapid releases of ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{129}I and ^{14}C from CANDU fuel were reported. For LWR fuel, there are far less data available /see e.g. Forsyth and Werme 1992, Gray et al. 1992, Gray 1999/.

It is generally assumed that the fission gas release and the rapid release of other segregated radionuclides are related. This seems to be a reasonable assumption for LWR fuel with gas releases in the range of a few percent /Johnson and McGinnes 2002/

For some radionuclides, assuming a one-to-one relationship seems to be a reasonable assumption, while for others the relationship seems to be more complex /Werme et al. 2004, Johnson et al. 2004/.

Radionuclide segregation to grain boundaries

In addition to the release of gaseous fission products, other elements that are incompatible with the structure of the uranium dioxide are also segregated to form separate phases. Light water reactor fuel contains inclusions of metal alloys of Mo-Tc-Ru-Rh-Pd (4d metals), known as ε-Ru phases. In normal fuel, these particles can be up to micrometers in size and observable by optical microscopy /Forsyth 1995/. The fraction of 4d metals, which Forsyth was able to determine by optical microscopy corresponded to only 1 percent of their calculated total inventory in the fuel. By electron microscopy, it has been possible to determine that ε-Ru phases also occur in the size range 1 to 100 nm /Thomas and Guenther 1989/. When fuel is dissolved in nitric acid, the 4d metals form an insoluble residue. By analysing these residues, Forsyth was able to conclude that at least 80 percent of the inventory of ε-Ru particles was present as spherical inclusions in the size range 0.1 to 0.5 microns.

Besides these known segregants the possibility that other fission products have segregated to the grain boundaries in the uranium dioxide has been discussed. For CANDU fuel, which is irradiated with higher linear power density (20–55 kW/m) than light water fuel, it has been established by photoelectron spectroscopy that Cs, Rb, Te and Ba are present in the grain boundaries, often with high surface enrichments /Hocking et al. 1994/. /Gray et al. 1992/ determined the grain boundary inventory of Cs in light water reactor fuel to be less than 1 percent. The values of gap and grain boundary inventories for Tc and Sr were near the detection limits at less than 0.2 percent of their total inventory. An Auger spectroscopy study on fuels with moderate burn-up and low fission gas release by /Thomas et al. 1988/ showed no detectable amounts of Cs, Sr and Tc. On a PWR fuel with extremely high fission gas release (18 percent; equivalent fuel was also included in the study by /Gray et al. 1992/), /Thomas et al. 1988/ found ε-phases with high surface concentrations of Cs, Te and Pd. This observation was interpreted as indicating that Cs and other fission products are mainly associated with the ε-phases and, thereby, indirectly with the grain boundaries that contain them. Since no detectable quantities of Cs, Sr and Tc could be detected in the grain boundaries, /Thomas et al. 1988/ drew the conclusion that the grain boundary inventory is small and can, therefore, hardly be regarded as a source of rapid release of these elements from the fuel.

The question of Sr segregation and the differences between Sr and U release from spent fuel in experiments at Studsvik has been discussed extensively. /Werme and Forsyth 1988/ offer the hypothesis that most of the strontium that is released comes from selective leaching of cracks and grain boundaries. Attempts have been made to determine grain boundary inventories by microprobe analysis in a fuel with a burn-up of 37 MWd/kg U (linear power density < 20 kW/m) that had been subjected to controlled power ramping to 43 kW/m in the Studsvik R2 reactor /Forsyth et al. 1988/. The rod had an appreciable release of Kr and Xe, as well as redistribution of Cs to the fuel-clad gap during the power increase. Sharp concentration gradients within the individual grains could be observed for Xe and Cs, but not for Nd. In the case of Sr, the concentrations were far too close to the detection limit to yield reliable data. In a subsequent corrosion experiment with a fuel with a burn-up of 44–48 kWd/kg U (linear power density < 15 kW/m), a similar power ramping to a peak of 43 kW/m was performed /Forsyth et al. 1994/. A significant increase in the release of Cs and Rb was observed, and some effect for Ba and Tc as well as possibly also for Mo. No significant redistribution of Sr was found.

Available data thus suggest that, in contrast to CANDU fuel, light water reactor fuel irradiated with moderate power densities has, at most, a few tenths of a percent of the inventory of radionuclides in the grain boundaries. Aside from the ε-Ru phases, there are probably no segregants in the fuel that would be of any significance. The grain boundary inventory is, within the margin of error, equal or close to zero. A possible contribution from the grain boundaries will be small compared to the inventory in the gap and can be modelled as included in the expected gap inventory for the fuel.

1.5 Definition of fuel variables

The spent fuel is described by the variables in Table 1-3, which together characterise the spent fuel in a suitable manner for the safety assessment. The description applies not only to the spent fuel itself, but also to the cavities in the canister.

The fuel and the cavity in the canister are characterised with respect to radiation by the intensity of α, β, γ and neutron radiation and thermally by the temperature. Hydraulically, it is necessary to characterise the cavity only if the copper canister should be damaged and water should enter. The cavity is then characterised by water flows and water pressures as well as by gas flows and gas pressures, which are jointly termed hydro-variables. Mechanically, the fuel is characterised by stresses in the materials, and chemically by the material composition of the fuel matrix and metal parts, as well as by the radionuclide inventory. The gas composition and, if water enters the canister, the water composition are also relevant for the description.

1.6 Summary of handling in safety assessment

Table 1-4 summaries the handling of fuel processes in the safety assessment SR-Can, as suggested in this report. In the table, the process is either “mapped” to a model by which it will be quantified or associated with a brief verbal description of how it will be handled.

Table 1-3. Variables for fuel/cavity in canister.

Variable	Definition
Geometry	Geometric dimensions of all components of the fuel assembly, such as fuel pellets and Zircaloy cladding. Also includes the detailed geometry, including cracking, of the fuel pellets.
Radiation intensity	Intensity of alpha, beta, gamma and neutron radiation as a function of time and space in the fuel assembly.
Temperature	Temperature as a function of time and space in the fuel assembly.
Hydrovariables (pressure and flow)	Flows, volumes and pressures of water and gas as a function of of time and space in the cavities in the fuel and canister.
Mechanical stresses	Mechanical stresses as a function of time and space in the fuel assembly
Radionuclide inventory	Occurence of radionuclides as a function of time and space in the different parts of the fuel assembly. The distribution of the radionuclides between the pellet matrix and surface is also described here.
Material composition	The materials of which the different components in the fuel assembly are composed, excluding radionuclides.
Water composition	Composition of water (including any radionuclides and dissolved gases) in the fuel and canister cavities.
Gas composition	Composition of gas (including any radionuclides) in the fuel and canister cavities.

Table 1-4. Process table for the fuel describing how fuel processes will be handled in different time frames and in the special cases of failed canisters. Green fields denote processes that are neglected or irrelevant for the time period of concern. Red fields denote processes that are quantified by modelling in the safety assessment. Orange fields denote processes that are neglected subject to a specified condition.

	Intact canister	Failed canister
F1 Radioactive decay (LW)	Thermal model	COMP23
F2 Radiation attenuation/heat generation (LW)	Thermal model	Neglected as long-term releases occur after period of elevated temperatures
F3 Induced fission (criticality) (LW)	Neglected since there will be insufficient amounts of moderator inside the canister prior to failure.	Neglected since the probability is negligibly small if credit is taken for the burn-up of the fuel
F4 Heat transport (LW)	Thermal model	Neglected as long-term releases occur after period of elevated temperatures
F5 Water and gas transport in canister cavity, boiling/condensation (LW)	Not relevant	Description in Main report, integrated with other relevant processes
F6 Cladding failure (LW)	Not relevant	Pessimistic assumption
F7 Structural evolution of fuel matrix (LW)	Not relevant	Neglected since burn-up sufficiently low
F8 Advection and diffusion (LW)	Not relevant	Description in Main report, integrated with other relevant processes
F9 Residual gas radiolysis/acid formation (LW)	Neglected since negligible amounts of corrodants are produced	Not relevant
F10 Water radiolysis (KS)	Neglected	Neglected except for fuel dissolution, see that process
F11 Metal corrosion (LW)	Not relevant	Pessimistic handling: a) No barrier function, all radionuclides instantaneously released upon water contact in COMP23 b) 1,000 years for complete corrosion if advective conditions in buffer
F12 Fuel dissolution (KS)	Not relevant	Modelled as constant, pessimistic dissolution rate in COMP23
F13 Dissolution of gap inventory (KS)	Not relevant	Pessimistic, instantaneous
F14 Speciation of radionuclides, colloid formation (KS)	Not relevant	COMP23
F15 Helium production (LW)	Neglected since the amount of helium produced will not increase the pressure inside the canister enough to affect its mechanical stability.	Not relevant
F16 Radionuclide transport (PS)		COMP23

1.7 Initial state of the canister in SR-Can

The following is an overview description of the initial state of the fuel, i.e. its state at the time of deposition. Additional information is given in the SR-Can Initial State Report /SKB 2006d/.

1.7.1 Description of cast iron insert and copper canister

The disposal canister for the spent nuclear fuel consists of an outer 5 cm thick copper shell with an insert made from nodular cast iron. The copper shell will act as the corrosion barrier in the repository, while the insert will provide the necessary stability to the whole waste package when exposed to the different mechanical loads that it may encounter during the one million year time period of the safety assessment. The weight and the dimensions of a disposal canister intended for BWR fuel can be seen in Figure 1-4. As can be seen in the figure, the insert contains individual channels for each fuel element. For BWR fuel, there are 12 channels in each insert, while for PWR fuel there will be only 4 channels.

The copper in the canister shell is oxygen free and will fulfil the specifications in /EN 1976:1988/ for the grades Cu-OFE (UNS10100) or Cu-OF1 with the following additional requirements: O < 5 ppm, P 20 – 70 ppm, H < 0.6 ppm and S < 8 ppm.

The material for nodular cast iron will fulfil the requirements in /EN 1563:1997/ grade EN-GJS-400-15U regarding mechanical properties.

A summary of the geometry for both the BWR and PWR versions of the canister is given in Table 1-5. The given values are those specified in the technical specifications.

5 cm copper

Estimated weight (kg):

Copper canister	7,400
Insert	13,600
Fuel assemblies (BWR)	3,600
Total	24,600

__Figure 1-4.__ Weights and dimensions for the canister for disposal of BWR fuel.

Table 1-5. Geometry of the assembled canister.

Canister	
Length:	4,835 mm
Diameter:	1,050 mm
Total weight (including fuel):	24,600 kg (BWR) 27,000 kg (PWR)
Copper tube	
Wall thickness:	50 mm
Inner length:	4.575 mm
Inner diameter:	953 mm
Lid thickness:	50 mm
Bottom thickness:	50 mm
Cast iron insert	
Length:	4.573 mm
Diameter:	949 mm
Channels for fuel assemblies	See Figure 1-5
Number:	12 (BWR) 4 (PWR)
Side:	160 mm (BWR) 230 mm (PWR)
Distance between sides:	50 mm (BWR) 150 mm (PWR)

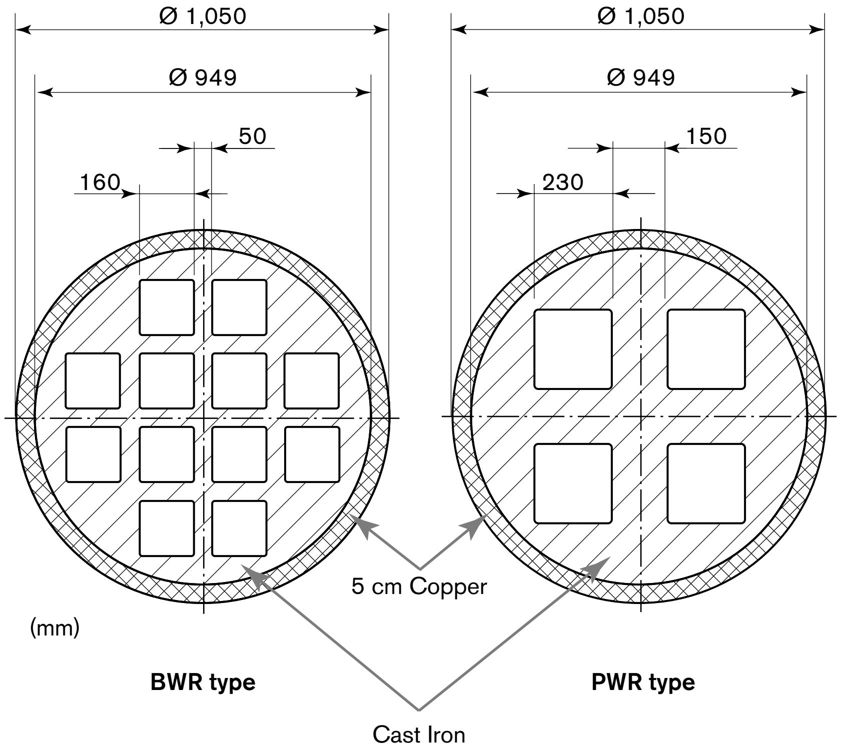

Figure 1-5. Dimensions of the cross section of the BWR and PWR canisters.

1.8 Canister performance and safety

In order to evaluate the canister's performance in the safety assessment, a number of so called function indicators and criteria that should be fulfilled over time, have been formulated, see further the SR-Can main report, /SKB 2006a/.

1.9 Definition of canister variables

The canister is described by the variables in Table 1-7, which together characterise the cast iron insert and the copper canister in a suitable manner for the safety assessment.

The fuel and the cavity in the canister are characterised with respect to radiation by the intensity of γ and neutron radiation and thermally by the temperature. Mechanically, the fuel is characterised by stresses in the materials, and chemically by the material composition of the cast iron insert and copper canister.

1.10 Summary of handling canister processes in SR-Can

Table 1-8 summaries the handling of canister processes in the safety assessment SR-Can, as suggested in this report. In the table, the process is either "mapped" to a model by which it will be quantified or associated with a brief verbal description of how it will be handled.

Table 1-6. Summary of canister safety function indicators and the criteria they should fulfil.

Function indicator	Criterion	Rationale	Reference
Copper coverage	$d_{\min}^{Cu} > 0$	Isolation is complete and no releases occur.	
Isostatic load	$P_{Isostatic}^{Canistrer/\,Buffer\,\mathrm{int}\,erface} < P_{Isostatic\,collapse}^{Can}$	Isostatic pressure in the buffer/canister interface should be smaller than the isostatic collapse pressure of the canister insert.	
Shear load	$RuptureLimit^{Canister} > ShearStress$	The rupture limit of the canister must exceed the shear stresses to which it is exposed	

Table 1-7. Variables for the cast iron insert and copper canister.

Variable	Definition
Radiation intensity	Intensity of gamma and neutron radiation as a function of time and space in the cast iron insert and copper canister.
Temperature	Temperature as a function of time and space in the cast iron insert and copper canister.
Canister geometry	Geometric dimensions of all components of the the cast iron insert and copper canister.
Material composition	The detailed chemical composition of the materials used for the cast iron insert and copper canister. This also includes cast iron and copper corrosion products.
Mechanical stresses	Mechanical stresses as a function of time and space in the cast iron insert and copper canister.

Table 1-8. Process table for the canister describing how canister processes will be handled in different time frames and in the special case of failed canisters. Green fields denote processes that are neglected or irrelevant for the time period of concern. Red fields denote processes that are quantified by modelling in the safety assessment. Orange fields denote processes that are neglected subject to a specified condition.

	Intact canister	**Failed canister**
C1 Radiation attenuation/heat generation (LW)	Included in integrated modelling of thermal evolution; thermal model	Neglected as long releases occur after period of elevated temperatures
C2 Heat transport (LW)	Included in integrated modelling of thermal evolution; thermal model	Neglected as long releases occur after period of elevated temperatures
C3 Deformation of cast iron insert (LW)	Isostatic: Comparison of external pressure with probabilistically calculated isostatic collapse load Uneven swelling: Neglect based on pessimistically simplified calculations and model calculations Tectonic events: Criterion for canister failure provided in process report. Creep for all above cases: Not included. Creep testing of the insert is currently in progress.	Not relevant
C4 Deformation of copper canister from external pressure (LW)	Initial creep due to buffer swelling pressure until gap closed Otherwise, deformation according to that of cast iron insert considering also creep	Not relevant
C5 Thermal expansion (both cast iron insert and copper canister) (LW)	Neglected since the thermal expansion will cause negligible strains in the materials	Not relevant
C6 Copper deformation from internal corrosion products (LW)	Not relevant	Description in Main report, integrated with other relevant processes
C7 Corrosion of cast iron insert (LW)	Not relevant	Description in Main report, integrated with other relevant processes
C8 Galvanic corrosion (LW)	Not relevant	Description in Main report, integrated with other relevant processes
C9 Stress corrosion cracking of cast iron insert (LW)	Neglected since stress corrosion cracking is considered unlikely and even if it occurred it would have no consequences for stability of the insert	Not relevant
C10 Radiation effects (LW)	Neglected since its effects on the mechanical properties if the insert would be negligibly low.	Not relevant
C11 Corrosion of copper canister (LW)	Sulphide in buffer and backfill modelled Microbially generated sulphide in buffer neglected if swelling pressure sufficiently high, otherwise pessimistically modelled (strictly a buffer process). Initial oxygen in buffer (strictly a buffer process): Pessimistically assumed that all oxygen corrodes copper neglecting oxygen consumption by buffer pyrite and rock? Initial oxygen in tunnel backfill (strictly a backfill process): Consider consumption by rock and microbes. Potentially intruding oxygen: Integrated handling of rock, backfill and buffer conditions. Pitting (oxygen corrosion): Described as uneven general corrosion Nitric acid corrosion: Neglected since only negligible quantities will be produced Chloride corrosion: Neglected since it requires very low pH to proceed under reducing conditions	Not relevant

	Intact canister	Failed canister
C12 Stress corrosion cracking, copper canister (LW)	Neglected due to the combined effect of very low (if any) concentrations of SCC promoting agents and the insufficient availability of oxidants.	Not relevant
C13 Earth currents – Stray current corrosion (LW)	Neglected due to e.g. the high polarisation resistance of copper under reducing conditions.	Not relevant
C14 Deposition of salts on canister surface (LW)	Process exists, but consequences negligible	Not relevant
C15 Radionuclide transport (PS)	Not relevant	COMP23

1.11 Handling of FEPs mapped to the fuel and canister process system

As a point of departure for the safety assessment SR-Can, a processing of features, events and processes, FEPs, of relevance for the long-term safety of a KBS-3 repository has been carried out. This work is further described in the SR-Can FEP report /SKB 2006c/.

The FEP processing has also provided lists of project FEPs in the international NEA FEP database version 1.2 that are related to the different geosphere processes. These FEP lists have been used by the experts developing the process descriptions, in order to ensure that all relevant aspects of a process are addressed in the process descriptions and appropriately handled in the SR-Can assessment. The handling of each FEP has been documented by the experts involved in developing the process descriptions in tables created for this purpose. These tables are provided in the SR-Can FEP report /SKB 2006c/ and the handling is also documented in the SKB FEP database.

In general, the results of these checks showed that FEPs were already covered by the process descriptions and included in existing plans for how to handle the process in the SR-Can assessment. In some cases, additional studies have been initiated as a result of the FEP analysis.

2 Fuel Processes

2.1 Radiation related processes

2.1.1 Radioactive decay

Overview/general description

The process of radioactive decay transforms the radionuclide content of the fuel and of those parts of the canister cavity to which radionuclides have spread. The radioactive disintegrations generate α-, β-, γ- and neutron radiation as well as new nuclides. These may also be radioactive and further decay until a stable nuclide is created.

This process is of fundamental importance, especially since it describes how the radiotoxicity and composition of the fuel evolves over time. The decay products may have chemical properties different from the parent nuclides that can affect their release mechanisms and migration properties. The energy liberated is converted for the most part into heat (Section 2.1.2), and the process thereby also constitutes the basis for the description of the repository's thermal evolution.

The following table shows how decay influences, and is influenced by, all fuel variables.

Boundary conditions

There are no boundary conditions of relevance for this process. (The initial condition is the radionuclide inventory in the fuel at the time of deposition of the fuel in the repository.)

Model studies/experimental studies

Radioactive decay has been thoroughly studied experimentally over a long period of time. The theoretical understanding of the process is good and fully sufficient for the needs of the safety assessment.

Time perspective

The time it takes for half of all radioactive atoms of a certain nuclide to decay is called the half-life of the radionuclide. The half-lives of various radionuclides vary from fractions of a second to millions of years.

Natural analogues/observations in nature

Radioactive substances occur naturally, and it is by studies of such natural analogues that the knowledge of radioactivity grew during the first few decades of the 20th century.

Handling in the safety assessment SR-Can

The process is included in integrated model calculations of the thermal evolution of the repository and of radionuclide transport (See Sections 2.2.1 and 2.6, respectively, for details regarding modelling).

Boundary conditions: As stated above, there are no boundary conditions of relevance for this process.

Handling of variables influencing this process: According to the influence table above, only the radionuclide inventory is of relevance. This variable is trivially included in the models of radioactive decay.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		Yes. The radiation intensity is a result of radioactive decay.	The radiation intensity is calculated from the radioactive decay of the inventory of radionuclides /see Håkansson 1999/.
Temperature	No		Yes. Most of the decay energy is transformed into heat.	The heat generation (and the temperature in the fuel) is calculated from the radioactive decay of the inventory of radionuclides (see above).
Hydrovariables (pressure and flows)	No		No Negligible in a failed canister compared to other processes influencing the hydrovariables	Neglected.
Fuel geometry	No		No	
Mechanical stresses	No		No, negligible	Negligible
Radionuclide inventory	Yes. This is the source of the process.	Obvious	Yes. The process both consumes and produces radionuclides.	The radionuclide inventory is calculated as a function of time (see above).
Material composition	No		No	
Water composition	No		Yes. Negligible compared to other processes influencing the water composition	Neglected.
Gas composition	No		Yes. Negligible compared to other processes influencing the gas composition	Neglected.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of the process is sufficient for the needs of the safety assessment.

Model simplification uncertainties

The radioactive decay as a function of time can be calculated with great accuracy when the nuclide content is known (See Sections 2.2.1 and 2.6 for details regarding modelling).

Input data and data uncertainties

The half-lives of the relevant radionuclides are generally known with good accuracy. Uncertainties concerning the inventory at deposition and half-lives are discussed in the SR-Can Data Report /SKB 2006b/.

2.1.2 Radiation attenuation/heat generation

Overview/general description

The radiation from radioactive decay interacts with the materials in the fuel and the canister cavity. Energy is thereby transferred to the materials and the radiation is attenuated. Most of the transferred energy is converted into thermal energy, i.e. heat is generated. The thermal energy, or heat, that is generated in the fuel after it has been taken out of service is called residual or decay heat and is dependent on the fuel's radionuclide content. This in turn depends on the fuel's burn-up and the length of the intermediate storage time before deposition in the repository. At first, the fuel's heat output is dominated by beta and gamma decay, but alpha disintegrations are also of importance in the long term /Håkansson 1999/. The isotopes which dominate during the first few centuries are Cs-137 (Ba-137m) and Sr-90 (Y-90), both with half-lives of around 30 years, resulting in a halving of the heat output in 30 years.

The process is of fundamental importance since it describes how heat is generated by the radiation. Furthermore, attenuation determines how much of the generated radiation reaches the canister.

The following table shows how the process influences and, and is influenced by, all fuel variables.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	Yes. The decay energy deposited in the fuel generates heat.	The heat generation is calculated from the radioactive decay of radionuclides /see Håkansson 1999/.	Yes. Attenuation lowers the radiation intensity.	Implicit in the model /see Håkansson 1999/.
Temperature	No		Yes	Obvious. Heat generation increases the temperature. Implicit in the model /see Håkansson 1999/.
Hydrovariables (pressure and flows)	Yes. In a failed canister, the presence of water will influence the radiation attenuation	Neglected. At the time of water intrusion, the γ-radiation will have decayed. α- and β-radiation will be attenuated by the fuel and the cladding.	No. Negligible in a failed canister compared to other processes influencing the hydrovariables	
Fuel geometry	(Yes) Most of the decay energy is deposited in the fuel and the geometry of the fuel is of subordinate importance.	Neglected	No	
Mechanical stresses	No		Yes. Increased temperature may create increased stresses in the fuel.	Neglected. This is of no importance for the stability of the fuel.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radionuclide inventory	Yes. The radionuclides are the source of heat.	Implicit in the model /see Håkansson 1999/.	No. The decay itself influences the radionuclide inventory. The generated heat does not.	
Material composition	Yes. Different materials have different attenuation properties.	Neglected. All fuel elements have essentially the same material composition	No	
Water composition	No		No	
Gas composition	No		No	

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundaries set by the geometries of the components involved and the loss of radiation over the boundary to the canister.

Model studies/experimental studies

The fuel's heat output is directly dependent on the radioactive decay process, as discussed in Section 2.1.1.

Time perspective

Heat generation in the fuel is only of importance for a few hundred years. After 1,000 years, heat generation is only about 5 percent of that at the time of deposition.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is modelled for the main scenario in SR-Can. The need for additional modelling in other scenarios will be evaluated.

The total power output of the fuel as a function of time has been calculated on the basis of its radionuclide content, decay energies and half-lives /Håkansson 1999/. For integrated modelling of the thermal evolution of the repository, this time dependent power output is approximated by a sum of exponential terms. All the heat generated is assumed to be deposited in the fuel rods.

Model: The thermal model is described in /Hedin 2004/.

Boundary conditions: Not relevant for this particular part of the integrated thermal model.

Handling of variables influencing this process: For the thermal modelling, all the radiation energy created in the radioactive decays is assumed to be deposited as heat in the fuel rods, yielding a simplified handling of all influences on the process listed in the influence table above. This handling is pessimistic with respect to the principal calculation end-point of the thermal model, namely the peak canister surface temperature, since no radiation energy is taken to escape the canister as gamma radiation.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of the process is sufficient for the needs of the safety assessment.

Model simplification uncertainties

Heat output as a function of time can be calculated with great accuracy when the radionuclide content is known. In the approximate handling through a sum of exponential terms, a sufficient number of terms are included for the approximation error to have negligible influence on the calculation end-point, the peak canister surface temperature. Also, the simplification of assuming that all heat generated is deposited in the fuel rods is pessimistic since, in reality, some energy will be deposited outside the canister and thereby has a smaller influence on the peak canister surface temperature.

Input data and data uncertainties

The uncertainties regarding heat output stem from uncertainties regarding the nuclide content of the fuel. These uncertainties are discussed in the SR-Can Data Report /SKB 2006b/.

Data uncertainties will be evaluated through sensitivity analyses.

2.1.3 Induced fission (criticality)

Overview/general description

Neutrons from radioactive disintegrations can cause nuclear fission in certain isotopes in the fuel. As long as the copper canister is intact, the great majority of neutrons generated by these disintegrations will pass out of the fuel without causing fission and the process can be neglected.

In the case of U-235 and Pu-239 in particular, the efficiency of the process increases if the neutrons are moderated (slowed down) to lower energies by collisions with light atomic nuclei. This could, to some extent, occur if water were to penetrate a failed canister. The water will then act as a moderator due to its content of light hydrogen nuclei. New neutrons are released by the fissions, and if more neutrons are formed than are consumed the process can become self-sustaining. The system is then said to be critical and large quantities of energy can then be liberated. It is this process that has been utilized under controlled forms in the nuclear reactor for energy production.

The following table shows how the process influences and, and is influenced by, all fuel variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	Yes. Neutron radiation is necessary for criticality to occur.	Neglected. The probability of criticality is considered to be negligibly small (see below).	Yes. Fissions will increase the radiation intensity.	Neglected. The probability of criticality is considered to be negligibly small (see below).
Temperature	No.		Yes. The fission energy will heat the fuel.	Neglected. The probability of criticality is considered to be negligibly small (see below).
Hydrovariables (pressure and flows)	Yes. The presence of liquid water is required.	Neglected. The probability of criticality is considered to be negligibly small (see below).	Yes. The heat generation may cause evaporation of water.	Neglected. The probability of criticality is considered to be negligibly small (see below).
Fuel geometry	Yes. Certain fuel geometries are more susceptible to criticality than other.	Neglected. The probability of criticality is considered to be negligibly small (see below).	No.	
Mechanical stresses	No.		Yes. Stresses due to increased temperature.	Neglected. The probability of criticality is considered to be negligibly small (see below).
Radionuclide inventory	Yes. The amount of fissile material as well as other nuclides in the fuel control the risk for criticality.	Neglected. The probability of criticality is considered to be negligibly small (see below).	Yes. Fissions lead to production of radionuclides.	Neglected. The probability of criticality is considered to be negligibly small (see below).
Material composition	(Yes) The amount of fissile material as well as other nuclides in the fuel control the risk for criticality.	Neglected. The probability of criticality is considered to be negligibly small (see below).	(Yes) Fissions lead to production of radionuclides.	Neglected. The probability of criticality is considered to be negligibly small (see below).
Water composition	No.		Yes. Increased radiolysis	Neglected. The probability of criticality is considered to be negligibly small (see below).
Gas composition	No		Yes. Evaporation of water and gaseous radiolysis products.	Neglected. The probability of criticality is considered to be negligibly small (see below).

In the repository, the spent fuel normal criteria for safety against criticality must apply. This means that the effective neutron multiplication factor k_{eff} including uncertainties must not exceed 0.95. /Agrenius 2002/ has calculated the effective neutron multiplication factor k_{eff} for a number of cases where the fuel is dry and water is in contact with the fuel. In nominal conditions the canister is leak-tight and the fuel is fresh. In this case with no water present the effective neutron multiplication factor is less than 0.5 and the system is indeed subcritical.

If it is assumed that the canister is leaking and that the canister storage positions and the fuel assemblies are water filled the reactivity will increase. With all storage locations in the canister filled with water the following results are found:

BWR: $k_{eff} = 0.9050 \pm 0.0012$

PWR: $k_{eff} = 1.0550 \pm 0.0012$

It can be concluded that the reactivity criteria could not be met with the conservative assumption that the fuel is fresh. A more realistic assumption would be to take credit for the burnup of the fuel, which will decrease the reactivity. /Agrenius 2002/ also calculated the neutron multiplication factor for irradiated fuel with various initial enrichments including the isotopic concentrations for three sets of isotopes:

Set 1: ^{234}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am.

Set 2: Set 1 + ^{243}Am, ^{237}Np, ^{143}Nd, ^{145}Nd, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{151}Eu, ^{153}Eu, ^{155}Gd.

Set 3: Set 2 + ^{99}Tc, ^{103}Rh.

Using Set 1 and and burn-up credit, the criteria $k_{eff} = 0.95$ could be met for both BWR and PWR fuel. Futher margin was obtained using Set 2, while Set 3 only yielded a marginal improvement.

The calculations by Agrenius were performed using Scale 4.4 /NUREG/. Depletion calculations were performed using the Scale SAS2H control sequence and the criticality calculations were performed using Scale CSAS25 (Keno V.a) sequence**.**

If the fuel assemblies have been damaged and uranium and plutonium have leached out, there are no probable courses of events that could lead to criticality /Oversby 1996, 1998/.

Boundary conditions

There are no relevant boundary conditions for the criticality process other than the physical boundaries set by the geometries of the components involved. (Other important factors for this process include the fuel burn-up, i.e. the fuel's content of fissile nuclides, and canister failure followed by the ingress of liquid water.)

Model studies/experimental studies

Nuclear fission has been thoroughly studied experimentally over a long period of time, and the theoretical understanding of the process is good and fully sufficient for the needs of the safety assessment. The issue of criticality in the canister has been studied by /Agrenius 2002/.

Time perspective

The fraction of fissionable material in the spent fuel changes insignificantly with time, but criticality in a failed, water-filled canister can be dismissed using the same analysis as discussed above.

Natural analogues/observations in nature

Criticality has been studied at the natural reactor in Oklo, see /Oversby 1996, 998/.

Handling in the safety assessment SR-Can

The probability of criticality is considered to be negligibly small, based on the results reported in /Agrenius 2002/ and in /Oversby 1996, 1998/.

Boundary conditions: As stated above, there are no relevant boundary conditions for this process other than the physical boundaries set by the geometries of the components involved. These are used in the reported studies of criticality.

Handling of variables influencing this process: All variables influencing criticality according to the influence table above are included in the reported studies of this process.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The process has been thoroughly studied within reactor physics. The mechanistic understanding is sufficient for the needs of the safety assessment.

Model simplification uncertainties

The reactivity of the fuel in the canister can be calculated with sufficient accuracy. Model uncertainties have been included in the assessment. A 2% uncertainty factor has been added to cover both bias and uncertainty in the calculational method. Statistical uncertainty in the Keno result was covered by adding an uncertainty of 0.004 Δk.

Input data and data uncertainties

Data uncertainties such as variation in burnup axially and horizontally, radionuclide content and manufacturing tolerances for the canister have been included in the assessment.

2.2 Thermal processes

2.2.1 Heat transport

Overview/general description

Heat is transported through the fuel and canister cavity by conduction and radiation to the canister insert and beyond to the near and far field. The process is dependent on the geometric configuration of the fuel and the thermal properties of the materials, which are given by the material compositions. The heat transfer to the canister insert sets the boundary conditions for the process. The result is a temperature change in the fuel/cavity.

The process constitutes a part of the thermal evolution of the repository.

The heat transfer between the fuel and the canister insert will through conduction and radiation. Which one of those that will dominate will depend on the composition of the residual gas in the cavity and the radiation properties[1] of the metal surfaces on the fuel assemblies and the canister insert. The temperature in the fuel and the cavity will be affected by the entire chain of heat transfers between the different components in the repository. In the longer perspective, the temperature evolution in the fuel and cavity will be controlled by the thermal power output

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[1] Expressed as emissivity, i.e. capacity to emit electromagnetic radiation.

of the fuel. In the shorter term, changes in the heat transfer properties of different repository components will have a greater influence on the temperature of the fuel. The peak temperature in the fuel will be reached within a few tens of years after deposition.

The following table shows how the process influences, and is influenced by, all fuel variables.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	No The radiation influences the temperature.		No	
Temperature	Yes Heat flow is controlled by the temperature and the temperature gradient.	The thermal evolution of the fuel and the canister is included in the integrated near-field evolution model /Hedin 2004/.	Yes Heat transport will increase the temperature at distances away from the heat source.	The thermal evolution of the fuel and the canister is included in the integrated near-field evolution model /Hedin 2004/.
Hydrovariables (pressure and flows)	Yes The gas pressure and the possible presence of water in the canister influence the heat transport in the canister cavity.	Included in the integrated near-field evolution model /Hedin 2004/.	Yes Increased temperature will increase the gas pressure.	Neglected.
Fuel geometry	Yes	Included in the integrated near-field evolution model /Hedin 2004/.	No	
Mechanical stresses	No		Yes Increased temperature may lead to heat induced stresses	Neglected.
Radionuclide inventory	No The decay influences the temperature.		No	
Materials composition	Yes Different materials have different thermal properties.	The thermal properties of the canister materials are included in the integrated near-field evolution model /Hedin 2004/.	No	
Water composition	No		No	
Gas composition	No		Yes The heat conduction depends on the type of gas in the cavity.	The thermal properties gas inside the canister are included in the integrated near-field evolution model /Hedin 2004/.

Boundary conditions

The heat transfer to the canister insert sets the boundary condition for the process.

Model studies/experimental studies

In the case of an electron beam welded copper canister, where there is a vacuum in the annulus between the copper shell and the cast iron insert, through which heat transfer will be controlled by the emissivity of the copper. The emissivity of electron beam welded copper has, therefore, been determined experimentally /Roos 2003/.

Hedin has developed a model for the thermal evolution of the fuel and the canister as a part of the integrated near-field evolution model for a KBS-3 repository /Hedin 2004/.

Time perspective

The peak temperature of the fuel is reached within a few tens of years after deposition. It then takes several thousand years for the fuel to cool off to near-ambient temperatures.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is modelled for the main scenario in SR-Can. The need for additional modelling in other scenarios will be evaluated.

The process is included in the integrated modelling of the thermal evolution, encompassing the fuel, the canister, the buffer and the host rock. The most important calculation end-point in the modelling is the peak canister surface temperature, which is insensitive to the handling of heat transport in the canister interior. To calculate the peak canister surface temperature, it is sufficient to know the power output as a function of time from the fuel. All other controlling factors are external to the canister.

Model: The thermal model described in /Hedin 2004/.

Boundary conditions: Heat transfer to the canister insert through both radiation and conduction is explicitly included in the model.

Handling of variables influencing this process: All variables influencing this process according to the influence table above are included in the model. The thermal conductivity in metal parts is assumed to be infinite, whereas heat transfer by radiation and conduction across gaps in the canister interior is handled for the explicit geometries of the fuel and canister.

Intruding water modifies the heat conduction properties in the interior of the canister only marginally. The description of heat conduction for the case of a defective canister is therefore assumed to lie within the frame of the uncertainties for the equivalent description for an intact canister. In addition, all conceivable causes of a canister failure are highly unlikely to occur during the fuel induced thermal transient in the repository.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of the process is sufficient for the needs of the safety assessment, especially since the calculation end-point of concern is insensitive to this process.

Model simplification uncertainties

The geometry of the model is simplified to a 2D-format, and heat conduction is neglected in some parts of the canister interior. These simplifications are either irrelevant or pessimistic.

Input data and data uncertainties

Uncertainties surrounding the heat transfer between the cladding tubes and the cast iron insert lead to uncertainties in the assessment of the temperature of the fuel. Compared with the uncertainties surrounding the heat transfer between the cast iron insert and the copper canister and between the canister surface and the bentonite buffer before full water saturation has been achieved, these uncertainties surrounding the heat transfer from the fuel to the canister insert are small.

As mentioned above, these data do not influence the calculation of the peak canister temperature.

Input data and data uncertainties are discussed in the SR-Can Data Report /SKB 2006b/. These uncertainties are explored through sensitivity analyses.

2.3 Hydraulic processes

2.3.1 Water and gas transport in the canister: boiling/condensation

Overview/general description

If the copper canister is penetrated, water can enter the canister cavity as liquid or water vapour. Transport of water, water vapour and other gases in the canister is then determined by the detailed geometry of the canister cavities, the presence of water/vapour in the cavities, and temperature and pressure. Boiling/condensation comprises an integral part of water/gas transport. Pressures and flows of water and gas across the container/buffer interface set the boundary conditions for this process.

The process is strongly coupled to several other processes; for example, corrosion of the canister insert, where water is consumed and hydrogen is formed. It is also of fundamental importance for some other processes that are dependent on the presence of water in the canister, such as fuel dissolution and radionuclide transport.

If the canister is penetrated, water will enter. The process is controlled by the pressure difference between the buffer and the canister cavity (typically 5–7 MPa) and the hydraulic conductivity in the bentonite. Water ingress can be expected to proceed very slowly. In the presence of water in the canister, gas will be generated both by radiolysis (see Section 2.5.3) and by corrosion of the cast iron insert (Section 3.5.1). The build-up of gas pressure in the canister will lead to a gradual decrease in the inflow of water and, when the pressure is sufficiently high, possibly also to gas transport through the canister and the bentonite. The process has been investigated by /Wikramaratna et al. 1993/ and /Bond et al. 1997/.

Qualitatively, the course of events can be described as follows: water enters through a penetrating defect in the copper canister and causes anaerobic corrosion of the iron insert surfaces. This leads to hydrogen gas formation, which gradually increases the pressure inside the cavity in the canister and thereby reduces the ingress of water. The corrosion leads to consumption of water and production of hydrogen gas. If the surface area available for corrosion is constant, the rate of water consumption will also be constant and the water level in the canister will reach a peak and then sink. (See also Section 3.5.1) Depending on the size of the hole in the canister and the corrosion rate, /Bond et al. 1997/ see three possibilities:

1) If the hole is so small that water is consumed by corrosion at the same rate as it enters, no water will collect inside the canister. The hydrogen gas pressure builds up gradually and approaches the external water pressure asymptotically. The corrosion rate will be governed by the inward diffusion of water vapour, which also declines asymptotically towards zero as the hydrogen pressure builds up.

The hole sizes for which this scenario applies depend on the corrosion rate. At a corrosion rate of 0.1 μm/y, the area of the hole must be less than about 8 mm^2 for the water pressure and bentonite conductivity assumed in the study. At a corrosion rate of 1 μm/y, (see also Section 3.5.1) the corresponding hole size is about 800 mm^2. When the inward transport of water due to the pressure difference between the inside and outside of the canister is lower than the inward transport by diffusion, canister corrosion will be diffusion-controlled (see below).

2) If the hole in the canister is larger than in case 1), but still sufficiently small, water will collect inside the canister. The water will be completely consumed before the pressure in the canister reaches the external pressure. Then the corrosion rate will be limited by the water supply rate, which will decrease with increasing hydrogen gas pressure leading to a decrease in corrosion rate (as in case 1). The hydrogen gas pressure gradually builds up and approaches the external pressure asymptotically, while the corrosion rate declines asymptotically towards zero.

In the same way as in case 1), the hole size defining this scenario is dependent on the corrosion rate. The combination of a corrosion rate of 0.1 μm/y and a hole size of 20 mm^2 belongs to this category, for example. When the inward transport of water due to the pressure difference between the inside and outside of the canister is lower than the inward transport by diffusion, canister corrosion will be diffusion-controlled (see below).

3) If the hole is sufficiently large in relation to the corrosion rate, the hydrogen gas pressure will reach the external pressure while there is still water in the canister. If the water level is then lower than the hole in the copper canister, hydrogen is expelled, but the internal pressure is still high and prevents new water from entering the canister. If the water level in the canister lies above the hole in the copper canister, water will first be pressured out until the water is on a level with the hole. Then hydrogen will be expelled as long as the corrosion continues, i.e. until all water in the canister has been consumed.

As in case 1, the hole size is dependent on the corrosion rate and can be illustrated by the cases 0.01 μm/y and hole size 5 mm^2, and 1 μm/y and hole size 0.18 m^2, both of which belong to case 3). (See also Section 3.5.1) When the inward transport of water due to the pressure difference between the inside and outside of the canister is lower than the inward transport by diffusion, canister corrosion will be diffusion-controlled (see below).

Even after all liquid water has been consumed, iron corrosion will continue since water vapour can diffuse into the canister. This case has also been modelled by /Bond et al. 1997/ for different corrosion rates, hole sizes in the canister, and available areas inside the canister for corrosion.

The continued corrosion will lead to a continuous increase in the hydrogen pressure. The hydrogen release from the canister will be controlled by the hydrogen pressure and the transport properties of the surrounding barriers.

The following table shows how the process influences, and is influenced by, all fuel variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes Influences the distribution between water and vapour	Neglected for an intact canister. For a failed canister the temperature is assumed to be constant at 11°C.	No	Neglected for an intact canister. For a failed canister the temperature is assumed to be constant at 11°C.
Hydrovariables (pressure and flows)	Yes Obvious	Neglected for an intact canister. For a failed canister the development inside the canister is based on the modelling by /Bond et al. 1997/.	Yes Obvious	Neglected for an intact canister. For a failed canister the development inside the canister is based on the modelling by /Bond et al. 1997/.
Fuel geometry	No		No	
Mechanical stresses	No		Yes Pressure build-up inside the canister	Neglected for an intact canister. For a failed canister, see 3.5.1
Radionuclide inventory	No		No	
Materials composition	No		No	
Water composition	Yes Water composition influences the corrosion rate	Neglected for an intact canister. For a failed canister see 3.5.1	Yes Release of soluble corrosion products	See 3.5.1
Gas composition	Yes Presence of water vapour	Neglected for an intact canister. For a failed canister see 3.5.1	Yes Hydrogen build-up	See 3.5.1

The transport of water and water vapour into the canister will lead to corrosion of the insert. The consequences of corrosion, such as the formation of soluble and solid corrosion products, and gaseous hydrogen will influence the water and gas transport inside the canister and, inversely, the products will influence the water and gas transport in the canister cavity.

Boundary conditions

Pressures and flows of water and gas at the canister/buffer interface set the boundary conditions for the process. The geometry of the damage in the copper shell forms an important part of these conditions.

Model studies/experimental studies

The process has been modelled for cracks in the canister by /Wikramaratna et al. 1993/, and in more detail by /Bond et al. 1997/.

No experimental studies of the process of water ingress and water consumption by hydrogen-generating corrosion have been carried out.

Time perspective

After canister penetration, the time frame is a matter of hundreds to tens of thousands of years /Bond et al. 1997/.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

For an intact canister, the water and gas transport in the cavity will be negligible from the point of view of long-term safety.

In the case of a canister failure, the process is treated in the SR-Can main report as a part of an integral description of the evolution of the canister interior after damage. The descriptions will be based on the modelling reported in /Bond et al. 1997/ and other sources. Other processes included in the integrated description are advection and diffusion, Section 2.5.1, corrosion of the cast iron insert, Section 3.5.1 and deformation of the canister from internal corrosion products, Section 3.4.5. Several alternative evolutions will be considered to account for conceptual uncertainties arising when integrating a number of processes. Gas release through the buffer must also be included in the description. This is discussed further in the Process report for the buffer and backfill, /SKB 2006g/.

The overall important effects of the coupled hydraulic and corrosion processes in the canister interior are *i*) a delay between the occurrence of the penetrating defect in the canister and the onset of radionuclide transport out of the canister and *ii*) an estimate of the evolution of the hole size as a function of time, which involves also mechanical processes. The purpose of the integral description is to arrive at pessimistic estimates of these two input parameters used in radionuclide transport calculations.

Model: If additional modelling is required, a similar model to that developed by /Bond et al. 1997/ is available as a module in the model described in /Hedin 2004/.

Boundary conditions: The boundary conditions mentioned above are explicitly included in the study by /Bond et al. 1997/.

Handling of variables influencing this process: All variables influencing this process according to the influence table above are included in the study by /Bond et al. 1997/. The temperature, however, is assumed to be constant and equal to 11°C and anaerobic conditions are assumed to prevail in the canister interior.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding is sufficient for a pessimistic treatment of this process in the safety assessment.

Model simplification uncertainties

The integrated internal evolution of a failed canister, of which this process forms a part, can be calculated with sufficient accuracy for the needs of the safety assessment, through pessimistic assumptions and by considering a number of scenarios. This is demonstrated in the SR-Can Main report.

Input data and data uncertainties

For the generation of corrosion gases, the corrosion rates are well-determined empirically and can be modelled with a constant corrosion rate if the water supply is not limiting /Smart et al. 2002ab/. If the corrosion rate is greater than the supply of water, the corrosion process will be controlled by the transport of water into the canister /Bond et al. 1997/.

Input data and data uncertainties regarding corrosion rates are discussed in the SR-Can Data Report /SKB 2006b/.

2.4 Mechanical processes

2.4.1 Mechanical cladding failure

Overview/general description

Temperature changes in the fuel and mechanical impacts associated with handling and transport will affect the fuel cladding.

Temperature increases lead to increased gas pressure inside the cladding tubes and can cause failure. Uptake of hydrogen during reactor operation may have led to hydride formation with embrittlement, which can lead to failure of the cladding tubes. The increased internal pressure may also lead to delayed failure through creep. In most cases, the damage will be local and result in a leak in the cladding tube.

The process is of importance for the release of radionuclides from the fuel. This can only happen if the cladding tubes are damaged.

The following table shows how the process influences, and is influenced by, all fuel variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes Causes increased gas pressure and increased creep rate.	Neglected. After failure of the copper canister all cladding tubes are assumed to be damaged.	No	
Hydrovariables (pressure and flows)	No		No	
Fuel geometry	No		No	
Mechanical stresses	No		Yes Increased mechanical stresses in the cladding due to temperature increase.	Neglected. After failure of the copper canister all cladding tubes are assumed to be damaged.
Radionuclide inventory	Yes Indirectly through heat generation.	Neglected. After failure of the copper canister all cladding tubes are assumed to be damaged.	No	
Materials composition	Yes Influence through the alloy used for cladding.	Neglected. After failure of the copper canister all cladding tubes are assumed to be damaged.	No	
Water composition	No		No	
Gas composition	No		No	

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundaries set by the geometries of the components involved.

Model studies/experimental studies

Experimental studies and modelling have been carried out for dry storage purposes /BEFAST III 1997/. Hydride formation and delayed failure have been studied for a long time /Northwood and Kosasih 1983, Grigoriev 1996/. /Rothman 1984/ conducted a review of the state of knowledge based on requirements for direct disposal of spent fuel. The conclusion was that failure due to hydride formation, though unlikely, cannot be entirely ruled out.

Time perspective

A small fraction of the fuel rods must be assumed to be damaged at the time of disposal. Even after disposal, hydride formation can lead to cracks in the cladding.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is of no importance for long-term safety as long as the copper canister is intact. To permit a discussion of retrieval, it can be pessimistically assumed that all cladding tubes may be damaged, but that they still provide physical protection for the fuel, and that the fuel assemblies retain their original geometry.

After failure of the canister, all cladding tubes will pessimistically be assumed to be damaged in the modelling of radionuclide transport.

Boundary conditions: As stated above, there are no particular boundary conditions to discuss for this process.

Handling of the variables influencing the process: With the pessimistic handling of this process, the variables influencing the process are also treated pessimistically.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The understanding of the process is not sufficient to permit reliable quantification. In cases where the process is of importance, i.e. when the copper canister is damaged, it will be handled pessimistically, as described above

Model simplification uncertainties

Not applicable since the process is not modelled.

Input data and data uncertainties

Not applicable since the process is not modelled.

2.4.2 Structural evolution of fuel matrix

Overview/general description

The time at which the fuel will contact water is estimated to be in the range of thousands to millions of years after deposition. The effects of radioactive decay, mainly alpha decay and build-up of helium in the fuel, could possibly change the physical and chemical behaviour of the fuel over these long time periods.

A redistribution of fission products due to thermally activated diffusion can be ruled out even for very long time periods /Werme et al. 2004/.

/Poinssot et al. 2000, 2001, 2002/ have analysed the possibility of alpha self-irradiation enhanced diffusion, also referred to as athermal diffusion. The analysis entails a scaling of the athermal diffusivity of fission gases due to fission-fragment irradiation to the value expected for irradiation by alpha particles. /Lovera et al. 2003/ have reported results of different theoretical approaches for studying the alpha self-irradiation enhanced diffusion. These models yield values after 100 years of decay ranging from $1 \cdot 10^{-25}$ to $1 \cdot 10^{-29}$ m^2/s. The diffusion coefficient is then expected to decrease with time as the alpha activity of the fuel decreases.

/Olander 2004/ was commissioned by SKB to investigate the possible consequences of such athermal diffusion on the long term stability of the instant release fractions of radionuclides. He analysed the thermal spike that is produced in the lattice as a result of alpha decay. It is this spike that would be responsible for athermal diffusion. The diffusion is actually thermally activated, but only on the local atomic scale in the vicinity of the alpha particle. The diffusion coefficient calculated for Xe as a result of the thermal spike integrated over all alpha decays was $1 \cdot 10^{-30}$ m^2/s, which is five orders of magnitude less than the highest diffusion coefficient reported by /Lovera et al. 2003/. The 10^{-25} m^2/s estimate chosen by /Lovera et al. 2003/ was obtained by assuming that all of the recoil energy resulted in a 10 nm permanent displacement from their lattice position of the atoms affected by collisions with the recoiling nucleus. This is an incorrect assumption since many of the atoms in UO_2 return to their lattice position after displacement /Van Brutzel et al. 2006/. In addition, for the 10^{-25} m^2/s estimate, it was assumed that no atom was affected by alpha recoil more than once. Thus, all recoil effects were assumed to move atoms from a lattice position to a non-lattice position. This is incorrect, since the likelihood of a collision with a recoiling nucleus is the same regardless of their location in the solid and the recoil can, therefore, lead to the relocation of an atom in a non-lattice position to a lattice position. Both of the invalid assumptions discussed above lead to overestimation of the diffusion coefficient due to defects caused by lattice damage from recoiling nuclei.

The athermal diffusion mechanism will lead to no significant contribution to release if the diffusion coefficient is less than $1 \cdot 10^{-26}$ m^2/s: For a diffusion coefficient of 10^{-26} m^2/s, the average movement of an atom, given by $x = (Dt)^{0.5}$, would be 5 nm in 100 years or 0.5 µm in 10^6 years. Therefore, for spent nuclear fuel of the types and burn-ups that are considered in SR-Can, athermal diffusion is not expected to lead to any increase of the instant release fractions even after a million years.

/Olander 2004/ used in his calculations PWR fuel with a burnup of 60 MWd/kgU and an out-of-reactor age of 20 yrs. His results gave an estimated diffusion coefficient for Xe of 10^{-26} cm^2/s (10^{-30} m^2/s). Since the diffusion coefficient increases due to alpha recoil increases linearly with alpha activity, the results from /Olander 2004/ show that no fuel that could possibly be generated in light water reactors could have a high enough alpha content to produce a diffusion coefficient due to alpha recoil that would be higher than 10^{-26} m^2/s.

The following table shows how the process influences, and is influenced by, all fuel variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	Yes. The alpha decay will cause radiation damage.	The process is neglected for the fuel types and burn-up that are relevant for a Swedish repository.	No	
Temperature	No. The temperature range will be too low to cause any annealing of any radiation damage.		No	
Hydrovariables (pressure and flows)	No		No	
Fuel geometry	No		No	
Mechanical stresses	No		No	
Radionuclide inventory	Yes. Indirectly through the content of alpha emitters.	The process is neglected for the fuel types and burn-up that are relevant for a Swedish repository.	Yes. If the process would be significant, the distribution of radionuclides in the fuel would be altered. For the fuel types and burn-ups considered in SR-Can, the effect is negligible.	The process is neglected for the fuel types and burn-up that are relevant for a Swedish repository.
Materials composition	No		No	
Water composition	No		No	
Gas composition	No		No	

The only variable of importance for this process is the total content of alpha emitters.

Boundary conditions

This is a diffusion process in the fuel rods, meaning that the boundary condition at the fuel rod surface is relevant.

Model studies/experimental studies

The process has been modelled by /Lovera et al. 2003/ and /Olander 2004/, see above. Experimental studies will not be possible because of the very low diffusion coefficients that are expected.

Time perspective

It is a matter of millions of years before the effect will contribute to the release from the fuel, if at all.

Natural analogues/observations in nature

Attempts have been made by /Lovera et al. 2003/ to apply their models for alpha self-irradiation enhanced diffusion to data from Oklo. The present relative composition of PbO in Oklo ranges between 5.4 and 7.7%. Theoretically, there should be 24.6%, i.e. substantial quantities of Pb have left the uraninite. If this loss was through diffusion, the diffusion coefficient would have been in the range of $7.5 \cdot 10^{-27}$ m/s^2 to $12.2 \cdot 10^{-27}$ m/s^2, i.e. within the range of diffusion coefficients proposed for alpha self-irradiation enhanced diffusion.

/Lovera et al. 2003/ ignored two important facts in their analysis. The Oklo uraninites that contain 5 to 8% Pb have been subjected to episodic loss of Pb on two occasions after their crystallization at the end of reactor operation. An intrusion of a swarm of dykes at 860 ± 39 Ma caused the expulsion of Pb from the uraninites. This Pb crystallized as mm-sized galena (PbS) crystals, which have an isotopic composition consistent with the Pb having grown in uraninite from 1,950 Ma to 850 Ma. The isotopic composition of Pb in the uraninites surrounding these galena crystals shows an initiation of growth of radiogenic Pb starting at 850 Ma and indicates that Pb was also lost from the uraninites at a later time. Detailed analysis of the isotopic composition data is consistent with loss of Pb at the time of the pan-African event, a regional thermal event, at about 500 Ma. Thus, the present Pb content of the uraninites is due to loss of radiogenic Pb during two high temperature events with relatively short duration, followed by ingrowth of new radiogenic Pb from about 500 Ma ago. The isotopic composition of Pb in the mm-sized galenas is consistent with negligible loss of Pb from the uraninites between 1,950 Ma and 850 Ma. The diffusion coefficient of Pb at low temperature from the uraninites during that time period must have been vanishingly small. For a complete discussion of the geochronology and petrography of the Oklo 5 to 6% Pb uraninites see /Evins et al. 2005/.

Handling in the safety assessment SR-Can

Based on the findings of /Olander 2004/, the process is neglected for the fuel types and burn-up that are relevant for a Swedish repository. The analysis of Olander was for spent fuel with 60 MWd/kgU burn-up and 20 years of cooling time, which represents less favourable conditions than those considered for spent fuels in SR-Can.

Boundary conditions: The process is internal to the fuel and there is no boundary, other than the zero concentration boundary condition on the fuel surface, included in Olander's model.

Handling of variables influencing this process: The only variable influencing this process is alpha activity, which decreases with time. This variable is trivially included in Olander's approach.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

Bounding estimates allow neglect of this process despite an incomplete mechanistic understanding (see above).

Model simplification uncertainties

Bounding estimates allow neglect of this process despite model simplifications, see above.

Input data and data uncertainties

Bounding estimates allow neglect of this process despite data uncertainties, see above.

2.5 Chemical processes

2.5.1 Advection and diffusion

Overview/general description

Solutes can be transported to and from the interior of the canister by advection and diffusion. These processes are not discussed explicitly, but dealt with integrated with other processes (often with pessimistic simplifications). (See also Section 2.6 for the handling of radionuclide transport).

The following table shows how the process influences, and is influenced by, all fuel variables.

The advection and diffusion in the canister cavity is influenced by the temperature, and require the presence of water or gas.

Boundary conditions

Pressures and flows of water and gas as well as concentrations of radionuclides at the transition to the buffer set the boundary conditions for the process.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes	For an intact canister, the process is neglected. In the case of a canister failure, the process is treated in the SR-Can main report as a part of an integral description of the evolution of the canister interior after damage.	No	
Hydrovariables (pressure and flows)	Yes	See above	No	
Fuel geometry	Yes	See above	No	
Mechanical stresses	No		No	
Radionuclide inventory	No		Yes The process redistributes radionuclides in the canister interior.	See above
Materials composition	No		No	
Water composition	No		Yes The process redistributes water solutes in the canister interior.	See above
Gas composition	No			

Model studies/experimental studies

There are no direct modelling or experimental studies for the case of a KBS 3 canister interior.

Time perspective

The process requires water intrusion. This is highly unlikely to occur before at least 1,000 years have elapsed after waste deposition in the repository, and, in the general case, before millions of years.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

For an intact canister, the process is neglected.

In the case of a canister failure, the process is treated in the SR-Can main report as a part of an integral description of the evolution of the canister interior after damage (see further Section 2.3.1). In the treatment of transport of dissolved species, the canister interior is essentially regarded as a well stirred tank.

For the case of radionuclide transport, see Section 2.6.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

Advection and diffusion are well understood mechanisms for solute transport.

Model simplification uncertainties

See Sections 2.3.1 and 2.6.

Input data and data uncertainties

See Section 2.3.1 and 2.6.

2.5.2 Residual gas radiolysis/acid formation

Overview/general description

Air and water in an intact canister can be decomposed by means of radiolysis. The products can then be converted to corrosive gases such as nitric and nitrous acids. These gases can be of importance for stress corrosion on the canister insert (see Section 3.5.3).

The following table shows how the process influences, and is influenced by, all fuel variables.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved.

Variable	Variable influence on process Influence present?	 Handling of influence	Process influence on variable Influence present?	 Handling of influence
Radiation intensity	Yes The intensity determines the extent of radiolysis	The amount of radiolysis products (nitric acid) is estimated based on the availability of nitrogen in the canister	No	
Temperature	No		No	
Hydrovariables (pressure and flows)	Yes The amount of radiolysis products depends on the availability of water and residual gases in the canister.	The amount of radiolysis products (nitric acid) is estimated based on the availability of nitrogen in the canister	No Any effect is negligible	
Fuel geometry	No Indirectly through the radiation intensity		No	
Mechanical stresses	No		No	
Radionuclide inventory	No Indirectly through the radiation intensity.		No	
Materials composition	No		No	
Water composition	Yes Dissolved solids and gases will influence the radiolysis products formed.	The water is assumed to be clean water with very low ionic strength.	Yes Radiolysis products will be formed in the water.	The amount of radiolysis products (nitric acid) is estimated based on the availability of nitrogen in the canister
Gas composition	Yes Gases will influence the radiolysis products formed.	The gas inside the canister is assumed to be at least 90% Ar and air.	Yes Gaseous radiolysis products will be formed.	The amount of radiolysis products (nitric acid) is estimated based on the availability of nitrogen in the canister

Model studies/experimental studies

The production of nitric acid has been calculated by /Marsh 1990/, /Henshaw et al. 1990/ and /Henshaw 1994/ for two different dose rates and for different quantities of air and water in the canister. Not unexpectedly, the quantity of nitric acid formed is dependent on the presence of sufficient quantities of water and air. The rate at which nitric acid is formed is dependent on the dose rate. The calculations show that with 50 g of water, i.e. the volume of water corresponding to the void volume inside a fuel pin, just under 160 g of nitric acid can be formed /Henshaw et al. 1990/. The availability of water will limit the amount of nitric acid that can form.

In the current plans for the encapsulation plant, the design goal is that no more than 600 g of water should be transferred to the canister with the fuel elements. In this case, the availability of water will no longer limit the amount of produced nitric acid. If the air is replaced with an inert gas, the quantity of nitric acid can be limited by the availability of nitrogen. During the encapsulation the air in the canister will be replaced by argon gas and at the most 10% residual air will remain in the canister. At that level of residual air, no more than 225 g of nitric acid can be formed.

Time perspective

The transformation of residual air and water to nitric acid takes place over the course of years to tens of years, /Henshaw et al. 1990, Henshaw 1994/.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

In an intact canister, residual oxygen, water and radiolytically formed nitric acid will be consumed by corrosion reactions with the canister insert. At the expected corrosion rates, it will take about 100 years to consume the oxidants inside the canister. As a consequence, all the oxidants will be consumed before a potential canister failure and will not have to be considered for a failed canister. The total depth of insert penetration by general corrosion is of the order of ten micrometres, which is negligible. The process is therefore neglected in the safety assessment. The consequences of stress corrosion cracking (SCC) are discussed in Section 3.5.3.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding is sufficiently good for the purpose of safety assessment.

Model simplification uncertainties

Mass balance considerations put an upper bound on the amount of nitric acid produced.

Input data and data uncertainties

The maximum allowed amount of water in a canister is 600 g. The uncertainty concerns the quantity of water that is actually inadvertently introduced into the canister. This quantity is expected to be much less than the allowed 600 g.

2.5.3 Water radiolysis

Overview/general description

Water that enters a damaged canister and reaches the cavities between the fuel assemblies and the canister insert is affected by the ionizing radiation emitted by the spent fuel. This causes the excitation, or ionisation of water molecules followed by breaking of their chemical bonds (*radiolysis*) producing primarily free electrons, OH-radicals and hydrogen atoms. Radicals are atoms or atomic groups resulting from the breaking of a chemical bond and containing a free or unpaired electron, which makes them highly reactive and able to participate in chain radiolytic reactions. Through such reactions with the solution components, a large variety of radicals is formed, as well as the stable molecular species O_2, H_2O_2, and H_2. Only gamma radiation, which has low Linear Energy Transfer (LET), and hence a relatively long range, is discussed here since it is the only form of radiation that can affect the water in the canister cavities. Radiolysis at the fuel/clad gap is dealt with Section 2.5.5, when fuel dissolution is discussed.

The production of reducing species by radiolysis results in a build-up of hydrogen gas in the void in the canister, while the simultaneous corrosion of the cast iron insert produces also hydrogen, as well as Fe(II) ions in solution. After some time, the hydrogen gas amounts produced by gamma radiolysis will reach a constant value and the result of continued gamma radiolysis will be reformation of water /Christensen and Bjergbakke 1982, Christensen and Bjergbakke 1984/.

The quantity of hydrogen gas produced by gamma radiolysis is dependent on the gamma dose rate. The contribution made by radiolysis to hydrogen gas formation is negligible compared with the contribution made by corrosion of the cast iron insert (see Section 3.5.1).

The following table shows how the process influences, and is influenced by, all fuel variables.

The radiation intensity affects the production of radical and molecular radiolysis products. At the time of water intrusion, the gamma field of the spent fuel is expected to have decreased to negligible levels.

The temperature affects the kinetics of radiolytic reactions and can be accounted for in radiolytic modeling. At the time of water intrusion into the canister, the temperature is expected to have decreased to near ambient values and the influence is expected to be negligible.

The presence of water in the canister is a requirement for the radiolysis process to take place, while the production of radiolytic gases affects the pressure in the canister. As discussed above, the effect on pressure is expected to be negligible in comparison to the production of hydrogen by the corrosion of iron.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	Yes The radiolysis is dose rate proportional.	Neglected In a failed canister the contribution to hydrogen gas production is considerably smaller than the contribution from iron corrosion. For radiolysis effects on fuel dissolution, see 2.5.5	No	
Temperature	Yes	Neglected See above.	No	
Hydrovariables (pressure and flows)	Yes Presence of water in the canister is required.	Neglected See above.	Yes Radiolytic gas production	Neglected See above.
Fuel geometry	No Indirectly through radiation intensity.		No	
Mechanical stresses	No		No	
Radionuclide inventory	No Indirectly through radiation intensity.		No	
Materials composition	No		No	
Water composition	Yes Radiolysis may form corrosive species. Water solutes participate in radiolytic reactions.	Neglected See above.	Yes Radiolytic production of water solutes.	Neglected See above.
Gas composition	Yes Dissolved gases participate in radiolytic reactions	Neglected See above.	Yes Radiolytic gas production	Neglected See above.

The fuel geometry affects the resultant gamma field inside the canister while the overall surface area of the fuel is of importance for the beta and alpha fields.

The influence of the radionuclide inventory is accounted for by the radiation intensity.

The composition of the water influences the yields and reactions of the different radiolytic species produced by radiolysis. The different radiolytic reactions in homogeneous solutions are modeled by kinetic radiolytic modeling with reliable results.

The presence of excess dissolved hydrogen in the groundwater solution inside the canister affects the gamma radiolysis of water through reactions of molecular hydrogen with radicals such as $OH\cdot + H_2 = H_2O + H\cdot$, which converts the oxidizing radical $OH\cdot$ to water and the reducing radical $H\cdot$ (atomic hydrogen).

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundaries set by the geometries of the components involved.

Model studies/experimental studies

Radiolysis of water has been thoroughly studied by both experiments and modeling. Low LET (Linear Energy Transfer) radiations, such as gamma, do not produce any detectable amounts of the stable species H_2 and H_2O_2 in pure deaerated water /Allen 1961/, because they react with $OH\cdot$ and $H\cdot$ radicals in a chemical chain reaction to reform H_2O. In aerated water virtually all the hydrated electrons and $H\cdot$ radicals formed are oxidized by O_2 and the main products of low LET water radiolysis in a system open to air are a steady state concentration of H_2O_2 with constant releases of H_2 and O_2 /Spinks and Woods 1990/.

Recent experimental and radiolytic modeling studies on the gamma radiolysis of water containing relatively small amounts of dissolved hydrogen /Pastina et al. 1999, Pastina and LaVerne 2001/ show that the production of oxidants involves a threshold, i. e. above a certain concentration of dissolved hydrogen no measurable oxidant production occurs. In studies of spent fuel corrosion in the presence of metallic iron /Loida et al. 1996, Grambow et al. 2000/ in 5 M NaCl solutions, no traces of radiolytic oxygen could be detected in the gas phase and reduced corrosion products of iron as magnetite and green rust, were analyzed on iron at the end of long term tests. The presence of such corrosion products on iron surfaces indicates an extremely low concentration of radiolytic oxidants in the solution. The absence of any radiolytic gas production in 5 M NaCl solutions containing 0.85 mM dissolved hydrogen was observed in another study /Kelm and Bohnert 2004/. The production of radiolytic gases in 5 M NaCl solutions was shown to restart in the presence of bromide ions.

Time perspective

The process contributes to hydrogen gas formation during the first centuries after deposition. After that, gamma radiation declines to negligible levels.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

In a damaged canister, the contributions to hydrogen gas production made by radiolysis and other effects are neglected, since they are considerably smaller than the corresponding effects of corrosion by water of the cast iron insert. The process can be neglected regardless of time for water intrusion, since the presence of Fe(II) and hydrogen from iron corrosion will result in negligible effects from radiolysis. In addition, gamma radiolysis is only relevant for a few hundred years after deposition in the repository.

Radiolysis effects on fuel dissolution is dealt with in Section 2.5.5.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of the process is sufficient for the needs of the safety assessment.

Model simplification uncertainties

Not relevant since the process is not modeled.

Input data and data uncertainties

Not relevant since the process is not modeled.

2.5.4 Metal corrosion

Overview/general description

The cladding tubes for the fuel are made of Zircaloy. Other structural elements in the fuel are made of stainless steel, Inconel, Incoloy or Zircaloy. Some of the radioactivity in the fuel is present in the cladding tubes and other metal parts as activation products.

If water enters the canister, the metal parts will corrode. The corrosion rate and the release of activation products from the Zircaloy will be controlled by the dissolution rate of the surface film of zirconium dioxide, which is tightly bound to the metal surface. The dissolution of the oxide film will be controlled by the solubility of ZrO_2 in the water in the immediate vicinity of the fuel and the removal of dissolved zirconium species. The solubility of ZrO_2 in water is very low, in the order of 10^{-9} M /Bruno et al. 1997/.

Considering the low corrosion rate of Zircaloy and in view of the low solubility of ZrO_2 in water, the rate of the release of activation products from the cladding tubes will be very low.

The following table shows how the process influences, and is influenced by, all fuel variables.

Any water in the canister cavity can lead to corrosion of cladding tubes and other metal parts in the fuel. The process is controlled mainly by the material composition, the chemical environment in the canister cavity, and the temperature.

The process affects the isolating function of cladding tubes and metal parts that enclose the fuel and is thereby of importance for the release of radionuclides from the fuel. The corrosion of metal parts also leads to release of the activation products that have formed in the metal parts.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	Yes Radiolysis may form corrosive species.	Neglected Release of all activation products from the cladding tubes is pessimistically assumed to occur immediately upon water contact.	No	
Temperature	Yes Corrosion rates tend to increase with temperature.	See above	No	
Hydrovariables (pressure and flows)	Yes Presence of water is required.	See above	No	
Fuel geometry	No		No	
Mechanical stresses	Yes May cause stress corrosion cracking of the fuel metal parts.	See above	No	
Radionuclide inventory	Yes Through the radiation intensity.	See above	No	
Material composition	Yes Different alloys may have different corrosion rates. The effect will most likely be negligible.	See above	No	
Water composition	Yes Metals have different corrosion rates in different waters. The effect will most likely be negligible	See above	Yes The influence is small due to the low Zr solubility.	See above
Gas composition	No		No	

Model studies/experimental studies

No studies of the corrosion rate of Zircaloy have been conducted by SKB, but a thorough literature review was performed in 1984 by the Lawrence Livermore National Laboratory for a tuff repository /Rothman 1984/. From Rothman's data, the corrosion rate can be estimated to be approximately 2 nm/y, i.e. penetration of the cladding tubes is estimated require 400,000 years at a tube thickness of 0.8 mm.

Although no detailed studies of corrosion of Zircaloy in the repository environment have been conducted, similar studies have been conducted for titanium /Mattson and Olefjord 1984, 1990, Mattsson et al. 1990/. The corrosion properties of titanium are very similar to those of zirconium; it is protected against general corrosion by a similarly adherent and insoluble surface film of titanium oxide. The results of the investigations showed that the oxide film remained tightly bound to the underlying metal and that the corrosion rate was extremely low at approximately 2 nm/y. This is very similar to the corrosion rate of Zircaloy based on Rothman's data.

Within the range of possible water compositions inside the canister, there will be no reason, within the accuracy of the estimated corrosion rates, to expect a dependence of corrosion rate on water composition and amount of water available as long as water or water vapour are present. Due to passivation of the Zircaloy, any radiolytically produced nitric acid is expected to preferentially corrode the cast iron insert.

The principal uncertainty is whether the corrosion rates observed in short-duration experiments are valid for geological time spans. The corrosion rate of Zircaloy (titanium) is measured during a short period compared with the expected life of the cladding tubes. The greatest uncertainty stems from the extrapolation of these short-term data. /Mattsson and Olefjord 1990/ derive a logarithmic growth law for the oxide layer on titanium for measurement data of up to three years. It shows a rapid growth of an initial passivating film of about 7 nm, after which further growth takes place extremely slowly. An extrapolation to 10^6 years gives an oxide layer thickness of 12 nm.

Data from /Mattsson and Olefjord 1990/ also show that for longer periods, 5 and 6 years, the growth rate deviates from the logarithmic law. After 6 years the film was 0.7 nm thicker than calculated, while the five-year samples showed a considerable distribution (from 4.9 and 15.5 nm) suggesting extrapolation according to a logarithmic law may not always be appropriate. For shorter exposures, the oxide film was amorphous, but after longer periods it was partially crystallized. It is, therefore, possible that the higher growth rate is attributable to a more rapid ion transport through grain boundaries. During the first year the film grows about 8 nm leading to the passivation of the freshly exposed surface. Consequently, a rate of 8 nm/y can be regarded as an upper limit for the corrosion rate and corresponds to a life of 100,000 years for the Zircaloy cladding.

The other engineering materials in the fuel are stainless steels or nickel-base alloys. SKB has not conducted any investigations of the corrosion resistance of these materials under repository conditions. /Gdowski and Bullen 1988/ have conducted a literature review of the corrosion resistance of similar candidate materials for encapsulation of nuclear fuel for the American Nuclear Waste Management Program. The corrosion rates for brief exposures in seawater, probably a more aggressive environment than the groundwater anticipated in a deep repository, suggest values of tens of microns per year for stainless steels and microns per year for nickel-base alloys. As with the maximum rate obtained for brief exposures of titanium (see above), these rates represent conservative upper limits, and long term rates will be considerably lower. The release of activation products will be controlled by these corrosion rates.

Since no relevant corrosion data have been gathered for other metal components in the fuel, the uncertainties are great. The corrosion rates are considerably greater than for the Zircaloy cladding, and a very pessimistic approach would be to set the life of these components at 0 years.

Time perspective

After water has come into contact with the cladding tubes, corrosion accompanied by release of activation products is expected to proceed at a constant rate for hundreds of thousands of years. The equivalent time frame for stainless steel components is hundreds of years, and for nickel-base alloys thousands of years.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

Available data suggest a life of the cladding tubes of at least 100,000 years. In SR-Can, however, the tubes are assumed not to have any barrier function against release of radionuclides from the fuel.

Release of all activation products from the cladding tubes is pessimistically assumed to occur immediately upon water contact.

Other metal parts are pessimistically considered to be immediately dissolved upon water contact as far as release and transport of radionuclides are concerned.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of the process is sufficient to allow a pessimistic handling in the safety assessment. Uncertainties are handled pessimistically since immediate release is assumed.

Model simplification uncertainties

Not relevant since the process is handled pessimistically without modelling.

Input data and data uncertainties

Not relevant since the process is handled pessimistically without modelling.

2.5.5 Fuel dissolution

Overview/general description

If water enters the canister cavity, the fuel may be dissolved/transformed, resulting in the release of uranium and other radionuclides contained in the fuel matrix. The process, which requires the protective cladding tubes have been breached, (Section 2.4.1), is controlled primarily by the chemical environment in the fuel-clad gap, particularly the presence of oxidants, and by the fuel composition, where matrix structure and the presence of radionuclides in the fuel matrix are decisive.

The process is of fundamental importance, since it describes the release of radionuclides from the fuel matrix and its contribution to the source term.

Non-matrix-bound activity

Radionuclides that have been segregated to the fuel-clad gap materials will rapidly go into solution. The quantity of activity released is determined by the solubility and availability of segregated material. The segregated radionuclides in the gap and grain boundaries usually have high solubility and mobility and are assumed to be released immediately upon water contact. The release from the gap is independent of the dissolution or transformation of the uranium dioxide matrix of the spent fuel and is discussed in more detail in Section 2.5.6.

Matrix-bound activity

The majority of fission products and higher actinides in the fuel exist as a solid solution in the uranium dioxide matrix. The release of these radionuclides requires that the uranium dioxide matrix of the spent fuel be dissolved or otherwise altered, for example by oxidation. This can happen only if water has entered the canister and the cladding tubes are breached. The redox conditions under which dissolution of spent fuel occurs are by far the most important factor influencing the dissolution process.

Redox conditions in connection with water ingress: When water comes into contact with the fuel, the uranium dioxide will begin to dissolve. This will occur by chemical dissolution of the $UO_2(s)$ matrix or by oxidative corrosion of the fuel, depending on the redox conditions in the immediate vicinity of the fuel. At the time of liquid water ingress, the corrosion of cast iron by water vapor is expected to have produced substantial amounts of hydrogen, part of which will dissolve in the intruding water and be present together with Fe(II) ions.

Any oxygen remaining in the repository near-field after closure should, at the time of groundwater ingress, have been consumed by bacteria and the reducing minerals in the rock and bentonite. Furthermore, the oxidation of iron by any intruding oxygen will likely proceed much faster than oxidation of UO_2 inside a damaged canister. An estimate of the time required to consume the oxygen in a copper canister shows that any residual oxygen will be consumed by oxidation of copper within a year or so /Johnson et al. 1996/. The consumption rate in an iron canister should be comparable, since the corrosion rates of iron and copper are comparable /Wersin et al. 1993/. There are, therefore, no appreciable uncertainties surrounding the assumption that the water that will come into contact with the fuel will be oxygen-free. Any contribution to the oxidative dissolution of fuel should, therefore, be caused by the radiolytic oxidants produced by water radiolysis.

The first entry of water into the canister is expected to occur more than a thousand years after deposition. By that time the predominant form of radiation will be alpha decay. Alpha particles have high LET (Linear Transfer Energy) and a very short range. They can cuase radiolysis of a thin layer of water (~35μm) near the fuel surface and produce mainly the molecular radiolysis products H_2O_2 and H_2. The effect of water radiolysis by alpha radiation on the dissolution of spent fuel has been studied by using solid UO_2 doped with ^{233}U. These samples contained alpha activity that approximated the levels expected 3,000 and 10,000 years after disposal of spent fuel. The tests were performed in the presence of anaerobically corroding iron. The results showed no evidence for enhanced dissolution of samples containing ^{233}U over those that contained only normal levels of ^{235}U and ^{238}U /Ollila et al. 2003/. The experiments were thoroughly analyzed for the location of precipitated uranium. Negligible amounts were found on the corroding iron /Ollila and Oversby 2005/. The release of U from the samples, especially during the first year or so of testing, appeared to be dominated by high energy surface sites formed by crushing of the samples or by high-energy interior sites at grain boundaries or associated with crystal imperfections. Similarly, low solution concentrations of U from ^{233}U doped samples in autoclave tests under hydrogen overpressure (corresponding to 3,000 years after disposal) have been reported by /Carbol et al. 2005/.

Since no enhanced dissolution due to alpha radiolysis was observed, there is no evidence of a mechanism to produce a redox front of oxidized radionuclides inside the canister. This is discussed further in the section Model studies/experimental studies.

The following table shows how the process influences, and is influenced by, all fuel variables. Further details are given below.

The process is influenced by radiation intensity, which determines the dose rate to water and the production of radiolytic oxidants. As discussed above, the dominating radiation at the time of water intrusion will be alpha radiation and its levels in spent fuel after 1,000 or more years are not expected to cause measurable oxidative dissolution of the uranium dioxide matrix in the presence of anaerobically corroding iron and its corrosion products /Werme et al. 2004/.

The temperature affects both the kinetics of all chemical reactions, including radiolytic and dissolution/precipitation processes, and the chemical equilibria in a damaged canister. At the time of water contact the temperature in the canister is expected to have decreased to near ambient and its influence is expected to be limited.

The presence of water in a damaged canister is required for the process to take place. Any potential beneficiary effect of secondary phase deposits on the rate of dissolution of spent fuel, e.g. by limiting the amount of water contacting the fuel surface, is pessimistically neglected.

The surface area of the fuel contacting water influences the dissolution rate, while the fuel geometry in the canister determines the resultant radiation field. Any influence of secondary precipitated phases on the fuel surface is pessimistically neglected, while any increase in fuel surface area due to dissolution of material from grain boundaries is considered negligible for the fuel dissolution rates assessed.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	Yes Affects radiolysis	Neglected The effect of radiolysis is assumed to be suppressed by the presence of H_2 and Fe^{2+}	(No) Small changes in inventory due to dissolution.	
Temperature	Yes Influences kinetics and equilibria.	Neglected Water ingress is assumed to take place at the earliest 1,000 years after disposal. At that time the temperature of the fuel will have a very limited effect.	No	
Hydrovariables (pressure and flows)	Yes Presence of water required for the process to take place	Water is assumed to be present after failure.	Yes Neglected influence of secondary phases on water flow is pessimistic	Pessimistically neglected
Fuel geometry	Yes Affects radiation field and radiolysis	Potential deposition of radionuclides on material surfaces.	Yes Changes in fuel surface area due to dissolution/ precipitation processes	Neglected
Mechanical stresses	No		No	
Radionuclide inventory	No Indirectly through radiation intensity		Yes Small changes due to release of radionuclides	Neglected
Material composition	No (Iron corrosion affects redox conditions indirectly through water composition.)		Yes Potential deposition of radionuclides on material surfaces.	Neglected
Water composition	Yes Affects radiolysis and kinetics of dissolution	Neglected The effect of radiolysis is assumed to be suppressed by the presence of H_2 and Fe^{2+}	Yes Soluble radionuclides from fuel dissolution	Source term for radionuclide release.
Gas composition	Yes Dissolved gases such as H_2 participate in radiolytic reactions	The effects of radiolysis is assumed to be suppressed by the presence of H_2 and Fe^{2+}	Yes Release of radioactive gases	^{14}C and Rn are assumed to be released with H_2 when it escapes from through the bentonite.

The radionuclide inventory of the fuel at the time of water intrusion is dominated by alpha emitting nuclides, since most of the short lived beta/gamma emitters have decayed after 1,000 years or more before water intrusion occurs. The fuel inventory of radionuclides determines the radiation field at the fuel surface and the dose rate to water as a function of time. The decrease of the inventory of a certain radionuclide in the fuel caused by its dissolution in water is usually very small.

The behaviour of the different materials in the canister, especially the anaerobic corrosion of the cast iron insert, influence fuel dissolution indirectly by changing the water composition and determining the redox conditions inside the canister, see Section 3.5.1. The composition of the different materials is not affected by fuel dissolution other than by the potential deposition of radionuclide precipitates on their surface.

The water composition influences both the radiolytic reactions and the kinetics of matrix dissolution. The most important factors are the redox potential and the concentration of carbonates, but other complexants, such as sulphate, chloride etc, affect the radiolytic reaction chains and the dissolution process.

The gas produced by the anoxic corrosion of iron influences the fuel dissolution process through changes in water composition. The process of fuel matrix dissolution causes the release of radioactive gases from fission gas bubbles as the dissolution progresses.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved. Related transport processes over the canister boundary are handled in Section 2.6.

Model studies/experimental studies

Experimental studies of fuel dissolution under various redox conditions have been conducted for more than 25 years. The majority of data in dilute groundwater conditions have been obtained under oxidizing conditions (see for example /Forsyth and Werme 1992, Forsyth 1997/). Several recent experimental studies have been carried out in the presence of various concentrations of dissolved hydrogen, varied over the range 0.05–43 mM, or in the presence of actively corroding iron. /Werme et al. 2004/ reviewed the available data under reducing conditions and proposed the fractional fuel dissolution rates to be used in the SR-Can safety assessment.

Recent experimental data on both spent fuel and alpha doped $UO_2(s)$ in the presence of H_2 and actively corroding iron indicate that the presence of a redox front or of a "chemical pump" process, during which the oxidized uranium released from the fuel surface is reduced and deposited on the surface of iron is highly improbable.

- More than 99% of the U(VI) released at the start of the leaching was deposited on the surface of the fuel itself /Albinsson et al. 2003/ and very low uranium levels were recovered in the vessel rinse.
- The transfer of relatively low U levels (40–200 ppb) present in solutions contacting metallic Fe under 10 bar H_2 pressure to a similar vessel containing spent fuel equilibrated during long time under the same conditions /Ollila et al. 2003/ caused a quick decrease of the uranium concentrations to much lower levels (a few ppb). This would indicate that spent fuel surfaces under such conditions have stronger reducing properties than Fe surfaces.

Similar observations were made during the simultaneous corrosion of spent fuel and iron /Grambow et al. 1996, 2000/. The uranium levels measured in solution were near the $UO_2(s)$ solubility, while the post characterization of Fe showed no $UO_2(s)$, even though magnetite and green rust were formed as corrosion products. Under oxygen free conditions it was shown recently that magnetite reduces U(VI) /Scott et al. 2005/, while green rust on iron surfaces was shown to precipitate $UO_2(s)$ /Cui and Spahiu 2002/. Apparently, during the co-dissolution of fuel and iron both oxidants and U(VI) were scavenged on the fuel surface and very little if at all on the iron surface. The same holds for leaching of ^{233}U doped $UO_2(s)$ in presence of iron extremely low U concentrations were measured in solution and negligible amounts of uranium were detected on iron surfaces /Ollila and Oversby 2005/.

Time perspective

After water has come into contact with the fuel, fuel transformation will proceed for hundreds of thousands of years.

Natural analogues/observations in nature

Not applicable.

In general, the conditions under which alteration occurred can not be accurately specified. In addition the alteration most probably occurred in the presence of oxygen. Therefore, there are no data on the alteration of uraninite that are relevant to repository conditions.

Handling in the safety assessment SR-Can

The fuel alteration/dissolution rate in the case of a damaged canister will be modelled using the recommendations in /Werme et al. 2004/ based on available experimental data from studies performed using alpha doped UO_2 and spent fuel under anaerobic and reducing conditions. These studies were performed in the presence of a hydrogen atmosphere or in the presence of actively corroding iron.

In the review by /Werme et al. 2004/, they conclude that the weight of the evidence from experimental studies suggests that the model for fuel dissolution in SR-Can should be a constant fractional dissolution rate in the range of 10^{-6} to 10^{-8} per year. A triangular distribution with a peak at 10^{-7} per year would best represent the available data. Support for this choice was given by /King and Shoesmith 2004/, who used an electrochemical model to predict the lifetime of spent fuel. They found that the maximum fractional dissolution rate in the presence of H_2 should be 10^{-7} to 10^{-8} per year.

Using this recommendation, the expected lifetime of the fuel is 10 million years with a distribution between 1 and 100 million years.

The recommendation of /Werme et al. 2004/ represents by far the most likely scenario, i.e. reducing conditions in the repository and the presence of hydrogen gas and/or corroding iron and iron corrosion products. Under these circumstances other parameters, such as water chemistry, will be of less important.

In the unlikely case of air saturated water coming into contact with spent fuel, the fuel will dissolve with higher rates and fractional dissolution rates in the range 10^{-4} to 10^{-5}/year, determined experimentally under oxidizing conditions, become appropriate /Forsyth and Werme 1992, Forsyth 1997/. In the presence of oxidants, the corrosion of the uranium dioxide causes an increase in concentration of U(VI) in solution followed by the precipitation of solid U(VI) phases on or near the fuel. The composition of these secondary phases will depend on the groundwater composition. In groundwater or bentonite pore water, uranyl oxides/hydroxides (schoepite) will probably form first, while, after longer times, more stable uranyl silicates (for example, uranophane or soddyite) will form.

Uncertainties

Uncertainties in mechanistic understanding

While there is a sound and increasing experimental database on the effect of the products of anoxic corrosion of iron on spent fuel dissolution, there are considerable uncertainties concerning the exact mechanism involved. This is also reflected in the wide spread in dissolution rates that /Werme et al. 2004/ propose.

Model simplification uncertainties

A constant fractional dissolution rate of the fuel is assumed in the calculations. The release of uranium and all other nuclides is assumed proportional to their inventory in the fuel matrix. The enhancement of radionuclides in the outer regions of the fuel matrix (the rim zone) is not considered since there are no direct observations that the rim dissolves faster than the rest of the fuel matrix /see e.g. Forsyth 1997/. In addition to this, the total volume of the rim zone is very small in comparison to the total fuel volume.

Input data and data uncertainties

Input data and input data uncertainties, i.e. the interval recommended by /Werme et al. 2004/ are discussed in the SR-Can Data Report /SKB 2006b/.

2.5.6 Dissolution of the gap inventory

Overview/general description

In the event of canister damage, water can enter the canister. If the fuel cladding is breached, the water can come into contact with the fuel. Most of the radionuclides in the fuel are evenly distributed in the UO_2 matrix and are released only when the uranium dioxide fuel matrix dissolves (see Section 2.5.5). A small fraction of the inventory of a few radionuclides has segregated to the fuel-clad gap and possibly also to grain boundaries in the fuel; see Section 1.4.4.

Most segregated radionuclides are also highly soluble and can quickly go into solution upon contact with water. The quantity of activity released is determined mainly by the availability and solubility of segregated material. A study of the release of segregated nuclides in fuel to be used in SR-Can has been performed by /Werme et al. 2004/. The release of this part of the inventory in a damaged canister is considered to occur instantaneously upon water contact.

The following table shows how the process influences, and is influenced by, all fuel variables.

The temperature influences the kinetics of the dissolution of the segregated radionuclides. At the time of water intrusion, the temperature is expected to have decreased to near ambient values.

The presence of water in a damaged canister is required for the process to take place.

The fuel surface area in contact with water influences the release of the segregated nuclides. This influence is accounted for in the estimation of the size of the instant release fraction.

The radionuclide inventory in the gap and grain boundaries determines the contribution of the segregated nuclides to the source term, while the dissolution of these radionuclides affects the radionuclide inventory of the fuel.

In principle, the composition of materials, such as fuel cladding, affects the dissolution of that part of the instant release fraction, which has diffused into the zirconium oxide layer during reactor operation.

Upon water contact, some segregated radionuclides, such as ^{14}C may produce gaseous products such as methane or carbon dioxide.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes	Neglected Instant release of the segregated inventory is assumed.	No	
Hydrovariables (pressure and flows)	Yes Presence of water necessary for the process	Water is assumed to be present after failure.	No	
Fuel geometry	Yes	Neglected Instant release of the segregated inventory is assumed.	No	
Mechanical stresses	No		No	
Radionuclide inventory	Yes Segregated inventory determines release	Instant release of the segregated inventory is assumed.	Yes Dissolution decreases radionuclide inventory	Instant release of the segregated inventory is assumed.
Material composition	Yes	Instant release of the segregated inventory is assumed.	No	
Water composition	Yes Limited influence of water composition due to fast release	Instant release of the segregated inventory is assumed.	Yes Small changes in composition due to limited amounts of segregated nuclides	Source term for radionuclide release.
Gas composition	No		Yes Gaseous products potentially formed.	Source term for radionuclide release.

Model studies/experimental studies

The immediate release of caesium and iodine from the fuel on contact with water is experimentally verified; see, e.g. /Johnson and Tait 1997/. /Johnson and McGinnes 2002/ have made a more recent assessment of the rapid release fraction from light water reactor fuel. An experimental study of the dissolution of segregated metallic ε-particles extracted from spent fuel /Cui et al. 2004/ under various redox conditions indicates very limited releases for the redox conditions expected in a damaged canister.

It has been experimentally shown that fractions of the content of the fission gases, Cs and I can leave the fuel matrix during reactor operation. It is also known that Tc, Ru, Rh, Pd and Mo form metallic inclusions in the fuel. There are no major systematic studies of segregants in light water reactor fuel, with the exception of the release of the fission gases. Uncertainties surrounding the extent, and in some cases the very existence of segregations of certain radionuclides are great. Uncertainties are discussed in greater detail by /Werme et al. 2004/.

Time perspective

The release of segregated radionuclides occurs relatively rapidly (on the order of days) and is pessimistically assumed to occur instantaneously upon water contact.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is only relevant if containment by the copper shell has been breached.

Immediate release of the rapid release fraction of radionuclides upon water contact in a damaged canister will be assumed for all scenarios in SR-Can. Input data will be based on distributions for the initial release fractions assessed in /Werme et al. 2004/.

Boundary conditions: As stated above, there are no relevant boundary conditions for this process other than the physical boundaries set by the geometries of the components involved. These become irrelevant with the pessimistic handling of the process.

Handling of variables influencing this process: the influences of temperature, fuel geometry, material composition and water composition are pessimistically neglected in the handling of this process. The influence of radionuclide inventory is trivially handled by assessing the fraction of segregated nuclides and assuming it is immediately released upon water contact. The requirement that water is present in the canister interior is handled by assessing a time after which a continuous water pathway has been established between the fuel and the canister interior.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of the process is sufficient for the needs of the safety assessment.

Any mechanistic/conceptual uncertainties are handled pessimistically since immediate release upon water contact is assumed.

Model simplification uncertainties

The uncertainties concerning the dissolution process are covered by the pessimistic assumption that the rapid release fraction of the radionuclide inventory is released immediately upon water contact.

Input data and data uncertainties

Input data and input data uncertainties for this process are discussed in the SR-Can Data Report /SKB 2006b/.

2.5.7 Speciation of radionuclides, colloid formation

Overview/general description

Radionuclides that have segregated from the fuel or are exposed to water in the fuel matrix and other metal parts can dissolve in the canister cavity and thereby become available for transport or precipitation when their solubility limit as an insoluble phase is reached. The distribution of a given nuclide between dissolved and solid phases is determined by the solubilities of the solid phases formed. These are in turn dependent primarily on the chemical environment in the canister cavity and the temperature. The low solubility of many key radionuclides is an important constraint limiting their concentration inside a breached canister and hence their rate of transport out of the canister. Radionuclide solubility refers to the total aqueous concentration of all the chemical species and isotopes of a radioactive element that are in equilibrium with each other and with a pure crystalline or amorphous solid phase containing the given element. If a true equilibrium with a characterized solid has been established, chemical thermodynamics allows the estimation of the maximum concentration of all soluble species for a given radionuclide in a specified groundwater composition. The solubility limited concentrations and chemical speciation of important radionuclides to be used in SR-Can have been reported in /Duro et al. 2005/.

Radionuclides can be released in the form of colloidal or pseudocolloidal particles from the spent fuel. Alternatively, colloidal particles may form during the nucleation of new solid phases inside the canister. This is of no importance for radionuclide dispersal from the canister, as long as the bentonite buffer, which acts as an efficient filter for colloids, remains in place.

The following table shows how the process influences, and is influenced by, all fuel variables.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	No		No	
Temperature	Yes	Neglected Water ingress is assumed to take place at the earliest 1,000 years after disposal. At that time the temperature of the fuel will have a very limited effect.	No	
Hydrovariables (pressure and flows)	Yes Presence of water necessary for the process	Water is assumed to be present after failure.	Yes	Effect of precipitates pessimistically neglected
Fuel geometry	No		Yes	Influence of solids pessimistically neglected
Mechanical stresses	No		No	
Radionuclide inventory	No		No	
Material composition	No Indirectly through water composition		Yes Deposition of precipitated solids on materials.	Modelled using solubility limits.
Water composition	Yes	Modelled	Yes Dissolved radionuclides in water	Source term for radionuclide release.
Gas composition	No Indirectly through water composition		Yes Dissolution of gaseous radionuclides.	Source term for radionuclide release.

The temperature influences both the kinetics of dissolution/precipitation processes as well as the solubility and complexation equilibria. For many solids, even the crystallinity of the solid formed depends on temperature; e.g. UO_2(s) precipitated from near neutral solutions at low temperatures is amorphous and the crystallinity increases with temperature /Rai et al. 2003/. At the time of a possible water intrusion into the canister, the temperature is expected to have decreased to near ambient values and its influence on solubility and complexation equilibria is expected to be small.

The presence of water in a damaged canister is necessary for the process to take place and the rate of water renewal affects the precipitation/dissolution process. In view of the large residence times of the groundwater in the canister and the relatively fast kinetics of inorganic ligand complexation and dissolution/precipitation reactions, the assumption of chemical equilibrium is not expected to introduce uncertainties in the calculation of solubility limited concentrations. Further, by assuming solubility equilibrium, maximum expected concentrations of a given radionuclide are obtained. Moreover, the relatively slow recrystallization of amorphous phases to more crystalline ones (Oswald's step rule) or the slow formation of solid phases as silicates is pessimistically neglected. The amounts of solids precipitated are not expected to be so large as to affect water flow or its presence in the fuel gap and any such influence on fuel dissolution is pessimistically neglected.

The radionuclide inventory does not in principle affect the precipitation or complexation processes, but determines the maximum availability for any given radionuclide at a given time.

Due to corrosion of the cast iron insert, reducing conditions are expected to exist inside the canister (see Section 3.5.1). Any oxidized forms of radionuclides released from the fuel will very probably be precipitated in their reduced forms by interaction with the iron corrosion products present on the surface of cast iron insert. This will give rise to a redox front inside the canister.

The composition of the groundwater inside the canister determines both the composition of radionuclide precipitates and their maximum soluble concentration. For redox sensitive radionuclides redox conditions in the groundwater are extremely important, and their variation can cause several orders of magnitude difference in the radionuclide concentration in some cases. Groundwater parameters, such as pH and the concentration of strong complexing ligands, e.g. carbonate, influence most of the equilibria, especially those of actinide and lanthanide elements. High salinity groundwaters affect the equilibria through their high ionic strength. Moreover, the dissolution/precipitation of radionuclides changes the composition of the groundwater inside the canister.

The presence of dissolved gases affects the redox conditions inside the canister but is not expected to influence solubility or complexation equilibria. The influence of pressures of the magnitude expected inside the canister on chemical equilibria is negligible /Marion et al. 2005/. On the other hand, certain radionuclides may enter the gas phase during the dissolution of either the rapid release fraction (Section 2.5.6) or the fuel matrix (Section 2.5.5) and establish a Henry's Law equilibrium.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved.

The presence of liquid water in a breached canister and sufficient progress of fuel dissolution to establish oversaturation with respect to a given element are necessary requirements for the process to take place. The chemical properties of the solution and the temperature influence the solubility of the potential solid phases.

Model studies/experimental studies

The quantitative estimation of radionuclide solubilities requires reliable thermodynamic equilibrium constants, obtained through experimental studies of appropriate chemical systems. The need for chemical equilibrium data in the field of nuclear waste management has contributed to a large number of basic thermodynamic studies on many radionuclides during the last 30 years. Most of this information has been reviewed by international expert teams within the framework of the NEA-TDB project /Grenthe et al. 1992, Silva et al. 1995, Rard et al. 1999, Lemire et al. 2001, Guillamont et al. 2003/.

In solubility calculations for a safety assessment, potential solubility-limiting phases are postulated for each radioelement. The most likely solid phases expected to form under the conditions that prevail are chosen. For example, amorphous phases are chosen instead of crystalline ones, while stable sulphides and silicates, whose formation is uncertain, are often disregarded entirely in the modeling.

The calculations determine the solubility of the radioelement phase in the groundwater in equilibrium with all the soluble species it forms under the given range of conditions, i.e. determine simultaneously the speciation of all radionuclides in the aqueous phase are determined. Speciation is important for both solubility, and the transport properties of the radionuclide in buffer and rock. The following methodology was used in deriving the solubility limits to be used in SR-Can /Duro et al. 2005/.

Solubility limits are defined for three different water compositions resulting from geochemical processes and climate changes. Special care has been taken to select a reliable and fully consistent thermodynamic database /Duro et al. 2004/. Chemical thermodynamic modeling has been used to assess solubility limits by varying the groundwater parameters including redox potential and pH for a range of pure solid phases which could potentially form under the expected conditions.

The output of the thermodynamic modeling is compared to literature data on the formation of identified minerals and measured concentrations in natural and laboratory systems. The extensive available spent fuel leaching data obtained under oxidizing or initially argon-purged atmospheres for important elements in the safety assessment have been discussed. A review of the solubility limits used in eight international safety assessment exercises has also been carried out /Duro et al. 2005/. Usually no credit is taken for the expected co-precipitation of elements with similar chemical properties, since the solubilities of the component radioelements in ideal solid solutions are lower than those of the corresponding pure components. A test case involving the calculation of radium-calcium-barium sulphate co-precipitation, for which there is abundant literature support /Doerner and Hoskins 1925, Berner and Curti 2002/, is the only exception.

Time perspective

If the canister is breached and water comes into contact with fuel, the process continues throughout the life of the repository.

Natural analogues/observations in nature

Elemental concentrations in natural systems as rocks, minerals or sea water as well as elemental concentration measurements in relevant natural analogue sites are discussed for most of the elements in /Duro et al. 2005/. The comparison of the results of thermodynamic modeling with data from natural and laboratory systems is used to check the consistency of the models with available experimental data.

Handling in the safety assessment SR-Can

The process is only relevant if the copper shell has been breached.

The process will be handled by applying concentration limits in the canister cavity water in the integrated radionuclide transport calculations.

Model: The near-field radionuclide transport model COMP23 /Cliffe and Kelly 2005/.

Concentration limits used in the calculations will be based on the results reported in /Duro et al. 2005/.

The solubilities and the speciation recommended in /Duro et al. 2005/ pertain to reducing conditions in the canister cavity, but for redox sensitive radionuclides, calculations for a range of redox conditions have also been carried out.

Boundary conditions: The process is confined to the canister and no precipitation of radionuclides in the form of solid phases is considered to take place in the buffer or in the far field.

Handling of variables influencing this process: The concentration limits applied are valid for a temperature of 15°C, which is a reasonable temperature for the canister interior after a few hundred years of deposition /Duro et al. 2005/. The influence of water composition is handled by calculating concentration limits for i) a site specific reference groundwater, ii) saline groundwater, iii) ice-melting groundwater and iv) a bentonite groundwater composition resulting from the interaction of the reference groundwater with bentonite.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The calculation of the solubility limits for each element of interest is based on equilibrium chemical thermodynamics and the fundamental understanding of the processes is sufficient for the needs of the safety assessment. Pessimistic assumptions are made whenever there are conceptual uncertainties. For example, all the uranium resulting from spent fuel dissolution is assumed to contribute to the ingrowth of radium and the uranium concentration in solution has not been limited by its low solubility under the reducing conditions expected to exist in the canister.

Model simplification uncertainties

The model does not account for any changes in water chemistry during the calculation.

Input data and data uncertainties

Input data and input data uncertainties for this process, i.e. the concentration limits to be used, are discussed in the SR-Can Data Report /SKB 2006b/, based on the data reported in /Duro et al. 2005/.

2.5.8 Helium production

Overview/general description

Helium builds up in spent fuel due to the alpha decay of actinides. Some of that helium may reach the gap between the fuel pellet and the Zircaloy cladding. This leads to a pressure build-up inside the tube, which can in turn lead to mechanical tube rupture (see Section 2.4.1). If, or when, the cladding ruptures, a negligible pressure increase arises in the canister cavity.

Most of the helium, however, will be trapped within the fuel matrix. This self irradiation and internal pressure build-up has the potential to detrimentally affect the mechanical stability of the UO_2 fuel matrix.

The following table shows how the process influences, and is influenced by, all fuel variables.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved.

Model studies/experimental studies

At 1,000 years after discharge from the reactor, ~~a~~ fuel with a burn-up of 35 MWd/kgU would contain about $5 \cdot 10^{18}$ He atoms/g, while a fuel with burn-up of 50 MWd/kgU would contain about $8 \cdot 10^{18}$ He atoms/g. At an age of 10^5 years, the concentration of He will have increased to $2.4 \cdot 10^{19}$ atoms/g and $3.1 \cdot 10^{19}$ atoms/g respectively. After a million years, the concentrations are less than two times higher than that.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	Yes The alpha decay rate determines the rate of helium build-up.	Neglected It is pessimistically assumed that all fuel rods are disrupted as soon as water contacts the fuel. The helium build-up in the canister void is neglected.	No	
Temperature	No		No	
Hydrovariables (pressure and flows)	No		No	
Fuel geometry	No		No	
Mechanical stresses	No		Yes Mechanical stresses in the cladding and possibly in the fuel.	Neglected It is pessimistically assumed that all fuel rods are disrupted as soon as water contacts the fuel. The helium buildup has no detrimental effect on the mechanical stability of the fuel
Radionuclide inventory	Yes Influenced by the content of alpha emitters.	Neglected See above	No	
Material composition	No		No	
Water composition	No		No	
Gas composition	No		Yes May result in He release.	Neglected Marginal amount in comparison to the H_2

A number of experimental studies of the possible consequences of helium build-up have been performed using He ion implantation /Desgranges et al. 2003, Roudil et al. 2004, Guilbert et al. 2003/. The implanted amounts were in the range $2{\cdot}10^{18}$ atoms/g to $2{\cdot}10^{20}$ atoms/g. Only in one case at $3.6{\cdot}10^{19}$ atoms/g was mechanical damage observed after heating to 500 and 600°C, but this may have been caused by the irradiation process /Werme et al. 2004/. It can be concluded that, for the burn-up range relevant for Swedish conditions, the mechanical stability of the spent fuel to be disposed of in Sweden will not be detrimentally affected by the accumulation of He in the fuel after disposal.

The amounts of He formed after 100,000 years (see above) correspond to 1.7 to 2.2 dm^3 (STP) He for each fuel pin. This is equivalent to a pressure rise of 3.4 to 4.4 MPa if released to the void volume (50 cm^3) in the fuel pin. In addition, the fuel pins may also have been pre-pressurised to a few MPa. If all the accumulated helium was released to the void inside the canister, the pressure rise would be 0.50 to 0.65 MPa (or about twice that amount if the fuel pins were pre-pressurized), after 100,000 years and about 1 to 1.3 after 1,000,000 years. This is considerably lower than the pressure external to the canister and will have negligible consequences for its stability.

Time perspective

The helium build-up proceeds continuously as long as there is uranium left in the fuel.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

It is pessimistically assumed that all fuel rods are disrupted as soon as water contacts the fuel, i.e. the ability of the cladding to prevent or retard radionuclide transport is pessimistically neglected.

The helium build-up in the canister void is neglected. The helium buildup has no detrimental effect on the mechanical stability of the fuel. If the helium is released to the void in the canister, the resulting pressure increase will not have any consequences for the mechanical stability of the canister.

Boundary conditions: As stated above, there are no relevant boundary conditions for this process other than the physical boundaries set by the geometries of the components involved.

Handling of variables influencing this process: The influence of the radionuclide inventory is explicitly included in all estimates justifying the neglect of this process.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

Uncertainties related to i) the fraction of the created alpha particles that actually contribute to helium pressure build-up, and ii) the impact of this pressure build-up on cladding integrity are treated pessimistically.

The main uncertainty related to the mechanical alterations of the fuel matrix is the uncertainty associated with future burn-up levels.

Model simplification uncertainties

The simplifications with respect to cladding barrier function and pressure build-up in the canister interior are pessimistic.

Input data and data uncertainties

There is some uncertainty considering the validity of short term He ion irradiation experiments for assessing the long term response of the fuel to He production. The He implantation procedure appears to disrupt the fuel matrix more than the actual alpha decay process.

2.6 Radionuclide transport

Overview/general description

If water enters a breached canister, corrosion of the cast iron insert and the fuel's metal parts begins, see Sections 3.5.1 and 2.5.4. With time, the evolution may allow water to come into contact with the fuel and thereby cause radionuclides to be released as water-soluble species or colloids (Section 2.5.7). Radionuclides are also present in the metal parts of the fuel and are released when they corrode.

Released radionuclides can be transported in the interior of the canister and can exit through a breached copper shell. Radionuclides dissolved in water inside the canister can be transported either advectively with the water, or by diffusion in the water. Transport in water is the predominant mode of mobilization of radionuclides. Some nuclides may occur in gaseous form in the canister and may then also be transported in the gas phase. This is particularly true of ^{14}C, ^{222}Rn and ^{85}Kr.

The geometry of the transport pathways in a breached canister is determined by the original geometry of the canister and the fuel and by the changes caused by corrosion has led. For water to come into contact with fuel and permit the release and transport of radionuclides, there must be penetrating breaches in both the cast iron insert and the Zircaloy cladding. Even if such breaches have occurred, the remaining structures can be expected to constitute considerable transport barriers, both to the inflow of water (Section 2.3.1) and to the outward transport of soluble nuclides. The structure of the four-metre-long cladding tubes, the other metal parts of the fuel, the cast iron insert and the copper canister prevent effective transport. Corrosion deposits from the cast iron insert in particular can be expected to obstruct transport. The surfaces of all of these structures may act as sorption sites for certain radionuclides.

The following table shows how the process influences, and is influenced by, all fuel variables.

Boundary conditions

The inner boundary is determined by radionuclide inventories in the fuel, and the release rate from the fuel and the concentrations of radionuclides in the water phase inside the canister.

The outer boundary is determined by size and shape of the defect in the copper canister.

Model studies/experimental studies

No direct studies of radionuclide transport are known for the conditions that are expected in a breached canister. The general knowledge of processes such as advection, diffusion and sorption is good, however, and the processes have been thoroughly studied.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes Diffusion is temperature dependent	Ignored Water ingress is assumed to take place at the earliest 1,000 years after disposal. At that time the temperature will have a very limited effect.	No	
Hydrovariables (pressure and flows)	Yes	Water is assumed to be present after failure.	No	
Fuel geometry	No		No	
Mechanical stresses	No		No	
Radionuclide inventory	Yes Obvious		No Obvious	
Material composition	No		No	
Water composition	Yes Affects solubility		Yes Dissolved radionuclides in water	Source term for radionuclide release.
Gas composition	No		Yes Release of gaseous radionuclides	Source term for radionuclide release.

Time perspective

After the copper shell has been breached, radionuclide transport in the interior of the canister is a relevant process in all of the safety assessment's time perspectives.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is only relevant if the copper shell has been breached.

Radionuclide transport in the interior of the canister is simplified for modelling purposes in the following way: After a given waiting period has passed since the breach of the canister's copper shell, the entire cavity initially available in the canister, about 1 m^3, is assumed to be filled with water. The length of the waiting period is determined based on the size of the breach in the copper shell and the subsequent water flux and corrosion.

After the waiting period, all the water in the canister is assumed to be available for the fuel dissolution process, i.e. to be in direct contact with all the fuel without being impaired by Zircaloy cladding or other structures. The water is assumed to be thoroughly mixed, i.e. there are no concentration differences throughout the interior of the canister. The fuel dissolution process then determines the rate of release of matrix-bound radionuclides. Segregated nuclides and radionuclides in the structural parts of the fuel are assumed to be available for dissolution in water immediately after the end of the waiting period. Sorption of radionuclides on the internal parts of the canister is neglected.

Transport of radionuclides through the breach in the copper shell is modelled as diffusion with an assumed geometry of the breach.

The modelling is carried out using the transport model COMP23 /Cliffe and Kelly 2005/, integrated with the modelling of fuel dissolution, precipitation/dissolution of solubility-limited radionuclides and radioactive decay. Transport through the buffer/backfill and out to the surrounding geosphere is also handled in the same model.

The micropores in the buffer are expected to prevent all colloidal transport out of the canister. Colloid transport in the interior of the canister is therefore not dealt with, since the buffer completely envelops the canister.

Radionuclide transport in the gas phase is handled by approximate calculations.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The uncertainties surrounding water/gas flux in the interior of the canister (Section 2.3.1), corrosion of the cast iron insert (Section 3.5.1) and corrosion of the metal parts in the fuel (Section 2.5.4) are particularly large. An assessment of the geometric conditions and thereby the geometry of the transport pathways is therefore also uncertain. The sorption properties of the materials in a damaged canister are also difficult to assess.

Model simplification uncertainties

Radionuclide transport in the interior of the canister is therefore simplified pessimistically in the safety assessment (see Handling in Safety Assessment).

Input data and data uncertainties

Input data and input data uncertainties for this process are discussed in the SR-Can Data Report /SKB 2006b/, based on the data reported in /Duro et al. 2005/.

3 Cast iron insert and copper canister

3.1 Radiation-related processes

3.1.1 Radiation attenuation/heat generation

Overview/general description

The gamma- and neutron radiation (from the radioactive disintegrations in the fuel) that extend into the canister interacts with the cast iron insert and the copper shell. Energy is thereby transferred to the materials and the radiation is attenuated. Most of the transferred energy is converted into thermal energy to generate heat.

In the cases of alpha and beta disintegrations, most of the radiation energy remains in the fuel since the radiation is largely attenuated by the fuel itself. In the case of gamma radiation, some of the surplus energy will escape the fuel and and generate heat by attenuation in the canister materials. This fraction is estimated to comprise less than 35 percent of the total heat output /see information given by /Håkansson 1999/.

The radiation attenuation in the canister will also have the potential to affect the mechanical properties of the canister materials. This is dealt with in Section 3.4.6.

The following table shows how the process influences, and is influenced by, all canister variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	Yes The radiation intensity causes the heat generation.	In the modelling of heat generation and dissipation, it is assumed that all heat generation takes place in the fuel itself.	No	
Temperature	No		Yes Radiation attenuation results in heat generation	In the modelling of heat generation and dissipation, it is assumed that all heat generation takes place in the fuel itself.
Canister geometry	Yes The degree of radiation will depend on the canister geometry, in particular the thickness of the various components.	The canister geometry and the relevant materials are explicitly included in the calculations leading to the neglect of this process.	No	
Material composition	Yes Different materials attenuate radiation to different degrees	See above		See Section 3.5.4
Mechanical stresses	No			See Section 3.4.4

The heat generation in the canister materials is caused by the radiation and, consequently, influenced by the radiation intensity and also by the canister geometry (metal thickness) and material. The attenuation/heat generation may also give rise to changes in the materials properties. This is dealt with in Section 3.4.6.

Boundary conditions

Part of the gamma- and neutron radiation from the radioactive disintegrations escapes the fuel into the canister and the energy will be attenuated by the canister materials. Most of the energy attenuation occurs in the inner parts of the canister. The remaining energy will cross the canister/buffer boundary and be dissipated outside the canister.

Model studies/experimental studies

Calculations of the canister's radiation attenuation (i.e. dose rate calculations) have been performed by /Håkansson 1995, 1999/ and /Lundgren 2004/. The dose rate in the channels in the canister was found to be about 300 Gy/h and at the outer surface of the canister was attenuated to less than 500 mGy/h.

Time perspective

Immediately after deposition, the gamma-radiation is dominated by decay of ^{137}Cs. This isotope has a half-life of approximately 30 years. In other words the radiation intensity is halved every 30 years.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

In the modelling of heat generation and dissipation, it is assumed that all heat generation takes place in the fuel itself. With this approach, the temperature in the fuel will be overestimated initially, but this will be of little importance for the temperature distribution in the near field shortly after deposition and beyond.

The process is not explicitly modelled, but the dissipated energy is accounted for. Section 3.4.6 discusses radiation effects on canister materials.

Boundary conditions: The relevant boundary conditions are explicitly included in the calculations leading to the neglect of this process.

Handling of variables influencing this process: The influencing variables, i.e. the radiation intensity, the canister geometry and the relevant materials are explicitly included in the calculations leading to the neglect of this process.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of this process is sufficient for the needs of the safety assessment.

Model simplification uncertainties

Not relevant since the process is not explicitly modelled in the safety assessment. (The model study used to justify the neglect of the process does not entail simplifications of any importance.)

Input data and data uncertainties

Not relevant since the process is not explicitly modelled in the safety assessment. (The available data are sufficiently accurate for the model study used to justify the neglect of the process.)

3.2 Thermal processes

3.2.1 Heat transport

Overview/general description

Heat is transported in the metal in the cast iron insert and copper canister by conduction. If electron beam welding is used there will be a vacuum in the gap between the cast iron and the copper and heat transfer in the gap will take place mainly by radiation. If friction stir welding is used, there will be an important contribution to the heat transfer from steel to copper from conduction since there will be air in the gap between cast iron and copper. There will also be a contribution due to heat conduction through the bottom contact between the insert and the copper canister.

The process is controlled by and influences the temperature distribution in the materials. The heat flow towards adjacent subsystems (fuel and buffer) comprises boundary conditions. The process is a part of the repository's thermal evolution.

The following table shows how the process influences, and is influenced by, all canister variables.

The radioactive decay (to which the radiation intensity is related) constitutes the heat source. The radiation intensity in the canister itself has negligible influence on the process. The transport of heat from the fuel is influenced by the canister geometry (metal thickness) and material. The temperature has a negligible influence on the heat transport through conduction, but a larger influence on the transport of heat through radiation.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	No The radioactive decay (to which the radiation intensity is related) constitutes the heat source.	The radioactive decay (to which the radiation intensity is related) constitutes the heat source.	No	
Temperature	Yes Obvious	The process is included in the integrated modelling of the thermal evolution, encompassing the fuel, the canister, the buffer and the host rock.	Yes Heats the canister materials.	The process is included in the integrated modelling of the thermal evolution, encompassing the fuel, the canister, the buffer and the host rock.
Canister geometry	Yes Obvious	See above	No	
Material composition	Yes Obvious	See above	No	
Mechanical stresses	No		Yes The increased temperature can induce thermal stresses in the material.	Neglected

The process is controlled by and influences the temperature distribution in the materials.

Due to the high thermal conductivity of iron and copper, very small temperature differentials arise within the metal components.

Boundary conditions

The boundary conditions are the heat flow from the fuel into the canister and from the canister out into the buffer.

Model studies/experimental studies

Temperature calculations have been carried out for the thermal evolution after disposal /Hedin 2004, SKB 2004b/. The calculations were carried out for a heat load of 1,700 W, which is assumed to be the highest heat load in a canister. The canister was assumed to be filled with air. The surface temperature of the outer copper shell was below 100°C.

Time perspective

Heat generation is halved every 30 years during the initial post-closure period. The highest temperatures and the greatest heat flows occur within tens of years after deposition and repository closure.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is modelled for the main scenario in SR-Can. The need for additional modelling in other scenarios will be evaluated.

The process is included in the integrated modelling of the thermal evolution, encompassing the fuel, the canister, the buffer and the host rock. The most important calculation end-point in that modelling is the peak canister surface temperature, which is insensitive to the handling of heat transport in the canister interior.

Model: The thermal model described in /Hedin 2004/.

Boundary conditions: The important calculation end-point, the canister outer surface temperature, is controlled by the heat generated as a function of time and by the thermal properties of the canister/buffer interface, of the buffer and of the host rock. Heat transfer due to both conduction and radiation at the boundary between the canister and buffer is included in the model. A case where the gap between canister and buffer is not symmetric, i.e. where the canister and buffer cylinder axes do not perfectly coincide may have to be considered.

Handling of variables influencing this process: All variables influencing this process according to the influence table above are included in the model. The thermal conductivity in metal parts is assumed to be infinite, whereas heat transfer by radiation and conduction across the gap between canister and buffer is included.

Heat transfer due to both radiation and conduction in gaps between the fuel and canister metal parts is included in the model. Heat conduction through the bottom contact between insert and copper canister is pessimistically neglected, since data are uncertain for this contribution and it only influences the calculated temperature of the fuel, of which a rough estimate is sufficient from the point of view of long-term safety.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The fundamental understanding of this process is deemed sufficient for the needs of the safety assessment.

Model simplification uncertainties

Model simplifications essentially involve a simplified geometry. The effects of such simplifications have been evaluated by benchmarking against complex models, which demonstrates that they have no significant influence on the calculation result; see further /Hedin 2004/.

Input data and data uncertainties

Assumptions regarding heat transfer between cast iron and the copper shell as well as heat transfer through the boundaries at the adjacent subsystems introduce the largest uncertainties.

Data relating to the thermal conditions in the gap between canister and buffer are important for the canister outer surface temperature. These, and other relevant data and data uncertainties, are discussed in the SR-Can Data Report /SKB 2006b/.

3.3 Hydraulic processes

Water and gas flux in the interior of the canister is described in Section 2.3.1 and will not be dealt with here.

3.4 Mechanical processes

3.4.1 Introduction

Mechanical interactions with the buffer occur at the boundary between the canister and buffer. They arise from the buffer through the clay matrix, which generates both compressive and shear stresses, through the pore water, which generates only compressive stresses, and through gas in the buffer, which also generates only compressive stresses. Changes in these three factors take place during the water saturation process and can also occur in response to external forces. The weight of the canister influences the buffer, while the influence of the weight of the buffer on the canister is negligible.

The mechanical interaction between buffer and canister involves the following:

- Swelling pressure and water pressure are exerted on the canister.
- Trapped gas in the buffer exerts pressure on the canister.
- Canister corrosion products exert pressure on the canister.
- Shear stresses transfer from the rock via the buffer to the canister, especially during an earthquake.

Swelling and water pressures are exerted on the canister

Swelling pressure develops as the buffer undergoes wetting. If the wetting is even, the swelling pressure increases evenly on the canister's periphery, except on the end surfaces where wetting of the buffer takes the longest time because of its thickness. If the wetting is uneven, the build-up of swelling pressure on the canister will be uneven. In the event of unfortunate combinations of wetting areas, the canister may be subjected to uneven stresses.

Water pressure on the canister does not arise until the bentonite is saturated. Theoretically, this pressure can also be uneven if certain parts of the buffer are saturated up to the canister while other parts are not.

The total pressure will be the sum of the swelling and the water pressures.
If the buffer is water-saturated before the maximum temperature has been reached, the water will expand a process which will be hindered by the confined volume. Thus, the pressure could theoretically be very large. However, such a pore pressure increase is compensated by water drainage out through the rock. Preliminary calculations show that no big pressure ever has time to build up /Pusch and Börgesson 1992/.

Trapped gas in the buffer exerts pressure on the canister

Gas generated or confined at the canister surface exerts a pressure on the canister. This gas pressure is, however, limited to the bentonite's opening pressure (see the SR-Can Buffer Process report, /SKB 2006g/). The gas is released through the bentonite either through temporary channels that form, or by dissolving in the water and diffusion out.

Canister corrosion products exert pressure on the canister

External corrosion of the copper canister, which can be both local and global, increases the canister volume, since the corrosion products have a lower density than copper. This increase causes consolidation of the buffer and an increase in the pressure between buffer and canister. Copper corrosion is, however, so slow that even corrosion over hundreds of thousands of years will not lead to more than a marginal increase in pressure, well within the error margins for the estimated buffer pressure on the canister throughout the one million year time period of the safety assessment.

In the event of a defect in the copper shell, a similar effect may occur in the cast iron. Preliminary modelling /Bond et al. 1997/ showed that the influence was not significant.

Transfer of shear stresses from rock via buffer to canister

Rock movements in the form of shear along a fracture plane affect the canister via the buffer. The buffer thereby acts to cushion the sharp and hard rock wedge. Since the movement does not change the total volume of the buffer, the effect is essentially a shear deformation, whereby the shear strength of the buffer regulates the impact. The shear strength is fairly low, since the friction angle at the density of the buffer is only about 10 degrees /Börgesson 1986/. The shear strength increases rapidly with increasing density, however, and doubles at a density increase of about 5 percent.

Thorough experimental and modelling studies have been made of the effects of rock shear on the canister and buffer. In model tests on a 1:10 scale, rock shear movements equivalent to approximately 20 cm shear in a deposition hole have been simulated /Börgesson 1986/. The effects on the buffer, copper shell and cast iron insert of faulting along a fracture intersecting a deposition hole has recently been modelled /Börgesson et al. 2004, Börgesson and Hernelind 2006, Hernelind 2006/. Laboratory tests were performed to determine relevant material properties for the buffer for the modelled situation /Börgesson et al. 2004/.

Shear rates of 1 m/s, buffer densities up to 2.1 g/cm^3 and net slips up to 20 cm were simulated. The results from the study indicate that the consequences of a faulting across a deposition hole depend strongly on the density of the bentonite, the type of bentonite (sodium or calcium), but also on the location of the fault in relation to the centre of the canister and the angle between the shear movement and the length axis of the canister.

3.4.2 Deformation of cast iron insert

Overview/general description

When the canister is loaded mechanically, for example when the buffer swells, stresses will be built up in the canister material, which deforms elastically at first but plastically if the stresses are large. The size of the deformation for a given load is dependent on the strength of the cast iron insert, which is determined by its material properties and geometry. At extreme loads the cast iron insert will collapse. Such loads, however, are much larger than any realistic load situation during the service life of the repository.

The cast iron insert is the most important mechanical barrier in the repository. If it collapses, a number of safety functions are jeopardized. The process is, therefore, of central importance to the function of the repository.

Under normal conditions in a geologic repository, the cast iron insert is expected to be subjected to an external load of approximately 18 MPa, composed of a maximum swelling pressure from the bentonite of around 13 MPa and a water pressure of 5 MPa. The pressure can be regarded as isostatic, i.e. evenly distributed over the entire surface. A canister can be subjected to an increased load during an ice age. An ice cover, several kilometers thick floating on the groundwater gives a maximum increase in pressure proportional to its maximum thickness. The expected pressure increase is about 28 MPa. The total load from these three causes can, thus, be estimated to be 45 MPa.

As the bentonite becomes water-saturated, uneven pressure distributions can arise. They may be associated with uneven water ingress or with oval or slightly curved deposition holes. In some cases an uneven stress could possibly persist for a long time. Such loads will, in addition to compressive stresses, also give rise to flexural stresses (tensile stresses).

Deviations from the normal load can also arise due rock displacements across the deposition hole. This could result in flexural and tensile stresses in the canister insert. Extremely large displacements (several decimetres) lead to canister failure.

A mechanical load also results from internal pressure build-up due to gas formation caused by alpha decay. The large void in the canister will make the effects of gas build-up negligible for all reasonable time periods (see Section 2.5.8).

If the copper canister is breached and water contacts the canister insert, it will corrode (see Section 3.5.1). The corrosion rate is low, but leads to a progressive reduction in the mechanical strength of the insert. This will eventually lead to failure. When and how this failure occurs depends on the extent of the breach, the geometry of the corrosion attack, and the load situation for the canister in question.

Creep deformation of the canister insert is not expected because the temperature will be low when loads are highest. Studies of the creep properties of cast iron are, however, in progress and it remains to be seen if this assumption is reasonable.

The following table shows how the process influences, and is influenced by, all canister variables.

Boundary conditions

The swelling pressure from the bentonite will control the development of even and uneven pressure situations.

Flat (subhorizontal) rock displacements across the deposition hole can cause an additional load on the canister.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No Discussed in 3.4.6		No	
Temperature	Yes The mechanical properties are temperature dependent.	Neglected The maximum mechanical load will occur after the period of increased temperature in the repository.	No	
Canister geometry	Yes The geometry and the properties of the material will determine the strength of the canister insert.	The canister's mechanical stability is modelled using the actual canister dimensions.	Yes A deformation will obviously alter the canister geometry.	Inherent in the model
Material composition	Yes The geometry and the choice of material will determine the strength of the canister insert.	The canister's mechanical stability is modelled using the actual canister materials properties.	No	
Mechanical stress	Yes Stresses are the direct cause of the deformation.	Inherent in the model	Yes A deformation will alter the mechanical stresses.	Inherent in the model

Model studies/experimental studies

Two tests with 700 mm long, full diameter canisters have been performed. In the first case, the test was performed on an insert that did not fully meet the specifications, while in the second test a "state-of-the-art" insert was used. In both cases, very high pressures were required for failure. Since the high-pressure press used for the test did not allow for continuous monitoring, the pressure was increased to a preset value. The pressure was subsequently relieved and the canister inspected for residual deformations before being subjected to a higher pressure. In the first test, pressures of 40, 70, 100 and 130 MPa were applied. The onset of deformation started at about 100 MPa, where a residual deformation of 5 mm was observed. At 130 MPa, the deformation was 20 mm. In the second case (loads of 40, 70, 100 and 130 MPa), a deformation of 5 to 13 millimetres was observed for a pressure of 130 MPa, while the insert collapsed at a pressure of 139 MPa /Nilsson et al. 2005/. These pressures exceed, by a large margin, the maximum expected load of around 45 MPa.

Since there, inevitably, will be a distribution of materials properties and quality among the inserts, a probabilistic analysis of canister stability has been carried out. This study, based on measured materials data from real canisters, showed that the probability for failure for the base case is vanishingly small (below : $2 \cdot 10^{-9}$). The same is also true if conservative assumptions are made both in the underlying deterministic analyses and the probabilistic analysis /Andersson et al. 2005, Dillström 2005/.

Uneven pressure build-up in the bentonite can occur both during and after water saturation. For the water saturation phase, three loading scenarios have been calculated using handbook load situations /Werme 1998/.

1) The canister is rigidly fixed at the end surfaces and along one-tenth of the length of the cylindrical surface nearest the end surfaces. An evenly distributed load, corresponding to a fully developed bentonite swelling pressure, acts along one side of the remaining canister surface.

2) The canister is supported at points situated one-tenth of the length of the cylindrical surface from the ends. An evenly distributed load corresponding to a fully developed bentonite swelling pressure acts along one side of the remaining canister surface.

3) The canister is rigidly fixed at one end surface and along one-fifth of the length of the cylindrical surface from that end. An evenly distributed load, corresponding to a fully developed bentonite swelling pressure, acts along one side of the remaining canister surface.

If the canister is tilted or inclined in the deposition hole, or if the rock is uneven, permanent pressure disequilibrium can arise in the bentonite even after water saturation with full water pressure and ice load. The following two cases have therefore also been calculated /Werme 1998/:

4a) The swelling pressure is fully developed on one side of the canister's cylindrical surface and on the end surfaces. On the other side of the cylindrical surface, the swelling pressure is 20 percent elevated along the central half and 20 percent reduced along the remaining quarters at the ends of the canister.

4b) Equivalent load cases, but where the swelling pressures are 50 percent higher and lower, respectively, are to be regarded as extreme cases for which no extra safety margins are required.

The results of the simple handbook calculations were that the yield strength of the cast iron insert (240 MPa, /Werme 1998/) is not reached in cases 1, 2, 4a or 4b, while case 3 would lead to collapse of the canister insert. The induced stresses are shown in Table 3-1. FEM calculations have therefore been carried out for cases 2 and 3, where the material properties of the bentonite have also been taken into account to give a more realistic load on the canister. The largest tensile stresses in the canister insert were then found to be lower than 55 MPa, which is considerably lower than the results obtained using the very simplified handbook calculations /Börgesson and Hernelind 1998/.

The buffer material in a deposition hole acts as a cushion between the canister and the rock, which reduces the effect of a rock shear substantially. Lower density of the buffer yields softer material and reduced effect on the canister. However, at the high density that is suggested for a repository the stiffness of the buffer is rather high. The stiffness is also a function of the rate of shear, increasing with increased shear rate, which means that there may be a substantial damage on the canister at very high shear rates.

In order to investigate the stiffness and shear strength of the buffer material a number of laboratory test series has been performed with shearing of water1saturated bentonite samples at different densities and shear rates. From those tests a material model of the buffer that takes into account the density and shear rate has been formulated. Shear rates up to 6 m/s have been tested /Börgesson et al. 2004/. The effects of faulting on buffer, copper shell and cast iron insert along a fracture intersecting a deposition hole was also subject to a first analysis

Table 3-1. Bending stresses for load cases 1–4.

Load case	Maximum bending stress (MPa)
1	122
2	183
3	690
4a	29
4b	71

/Börgesson and Hernelind 2006/ have made new calculations using an improved finite element model. The finite element code ABAQUS was used for the calculations. The calculations include both sodium and calcium bentonite as buffer materials. The rock shear movements considered are shear movements at 90° angle to the length axis of the canister and either at the middle of canister or at ¼ of the canister length. The calculations also include cases where the angle of the shear movement was 22.5° and 45° to the length axis of the canister and through the middle of the canister. This gives rise to two distinct cases for each shear angle. They are referred to as the compression and tension modes. In the compression mode, the shear movement will result in a reduction of the height of the deposition hole, while the tension mode will result in an increase of the height of the deposition hole. The two shear modes are illustrated in the sketch in Figure 3-1. The shear plane was assumed to go through the centre point of the canister.

The analysis by /Börgesson and Hernelind 2006/ provides the stress and plastic strain of the copper shell, the insert and the buffer immediately after shearing. A summary of the results is given in Table 3-2 for the case of sodium bentonite and in Table 3-3 for the case of calcium bentonite. The tables show the maximum plastic strain in the copper tube (excluding the lid) and in the cast iron at two different slips (10 and 20 cm).

The current specifications for the copper canister state that the copper material must have a ductility of at least 30 percent. For the cast iron insert, the ductility must be at least 7 percent. As can be seen in the tables, the canister will resist a shear movement of 10 cm with sufficient margin for densities up to 2,050 kg/m^3. For a rock displacement of 20 cm, the ductility limit of the cast iron insert is exceeded for the centric and eccentric cases for calcium bentonite with a density of 2,050 kg/m^3. The strain levels in the copper canister lid and bottom were locally higher than in the copper tube. The largest plastic strain found in this study was 19%. This is still within the ductility limit of the copper material.

The current specifications for the copper canister state that the copper material must have a creep ductility of at least 10 percent. /Hernelind 2006/ has supplemented some of the calculations by /Börgesson and Hernelind 2006/ by including creep relaxation of the stresses that were introduced into the copper shell. For the case of 2,000 kg/m^3 bentonite the resulting creep strain was 7.6 percent for a 0.1 m shear movement and 11.5 percent for a 0.2 m shear movement. With reasonable safety margins, the canister will withstand a 10 cm shear movement horizontally across the deposition hole. For a 0.2 m shear movement, creep failure of the copper shell cannot be totally excluded based on the results obtained with the current creep model.

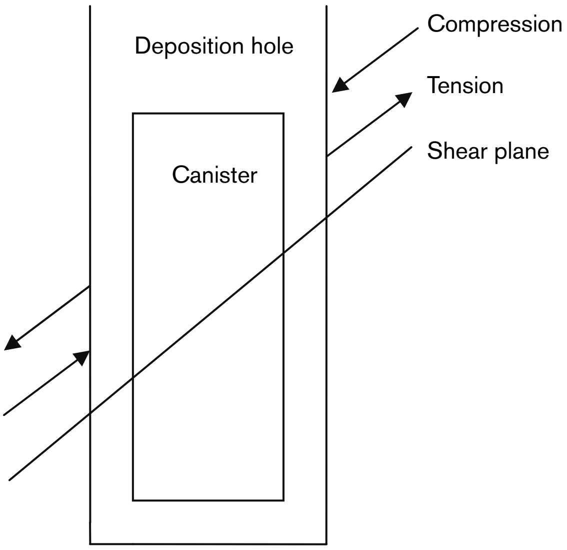

Figure 3-1. Illustration of the compression and tension shear modes.

Table 3-2. Calculated maximum plastic strain in the copper tube and in the cast iron insert at rock displacement of 10 and 20 cm (eccentric = shear at ¼ of the canister length and centric = shear at the middle of the canister). Sodium bentonite.

ρ_m (kg/m^3)	Shear angle (°)	Shear direction/ location	Maximum plastic strain (%)			
			After 10 cm shear		After 20 cm shear	
			Cu-tube	Fe-insert	Cu-tube	Fe-insert
2,000	22.5	Compression	0.2	< 1	0.3	< 1
2,000	22.5	Tension	2.5	< 1	3.3	< 1
2,050	22.5	Compression	0.3	< 1	0.4	< 1
2,050	22.5	Tension	3.5	< 1	4.6	< 1
2,000	45	Compression	0.5	< 1	0.6	< 1
2,000	45	Tension	3.5	< 1	5.0	< 1
2,050	45	Compression	0.8	< 1	1.5	< 1
2,050	45	Tension	5.5	< 1	8	< 1
2,000	90	Centric	0.2	< 1	0.3	< 1
2,000	90	Eccentric	1.2	< 1	2.0	1.7
2,050	90	Centric	0.4	< 1	0.5	< 1
2,050	90	Eccentric	1.2	1.3	2.5	3.6

Table 3-3. Calculated maximum plastic strain in the copper tube and in the cast iron insert at rock displacement of 10 and 20 cm (eccentric = shear at ¼ of the canister length and centric = shear at the middle of the canister). Calcium bentonite.

ρ_m (kg/m^3)	Shear angle (°)	Shear direction/ location	Maximum plastic strain (%)			
			After 10 cm shear		After 20 cm shear	
			Cu-tube	Fe-insert	Cu-tube	Fe-insert
2,000	22.5	Compression	0.4	< 1	0.6	2.8
2,000	22.5	Tension	4.4	< 1	6	< 1
2,050	22.5	Compression	1.0	3.6	2.5	11
2,050	22.5	Tension	6.4	< 1	10	1.5
2,000	45	Compression	1.5	< 1	2.0	2.9
2,000	45	Tension	7.5	< 1	11	1.4
2,050	45	Compression	4.5	1.7	10	5.9
2,050	45	Tension	10	< 1	19	4
2,000	90	Centric	1.2	< 1	2.5	1.5
2,000	90	Eccentric	2.5	2.3	6.5	4.9
2,050	90	Centric	7.2	5.4	16	13
2,050	90	Eccentric	5.5	2.4	13.5	7

Time perspective

The load from the groundwater pressure and the bentonite swelling pressure is expected to build up within a few years to a few decades after deposition. The maximum glacial load can be expected after tens of thousands of years.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

Isostatic load: The isostatic load on the canister over the one million year assessment period will be assessed in SR-Can. This load will be compared to the isostatic collapse load that can be estimated from the experimental and model studies cited above. The comparison will be used to assess whether canister integrity may be jeopardised due to isostatic over pressure.

Uneven pressure build-up in the bentonite: The effects of this load situation are neglected, based on the calculations in /Werme 1998/ cited above.

Effects of rock shear: The likelihood of violating the criteria for rock shear movements that the canister can sustain given above will be assessed, based on estimated rock and buffer conditions. If violation occurs, the canister integrity will be assumed to be lost.

Creep effects in the cast iron insert: For the temperature range in question, creep is expected to be negligible for all the load cases above (see Overview/general description above).

All these load situations will also imply loads on the copper shell. The effects on the copper shell are discussed in Section 3.4.3.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

Isostatic load: The fundamental understanding is sufficient for the needs of the safety assessment.

Uneven pressure build-up in the bentonite: The fundamental understanding is sufficient for the needs of the safety assessment.

Effects of rock shear: The fundamental understanding of the response of the canister insert to a rock shear movement is sufficient for the needs of safety assessment.

Model simplification uncertainties

Isostatic load: There are no model simplification uncertainties of any importance for the safety assessment.

Uneven pressure build-up in the bentonite: The uneven load cases shown in Table 3-1 are modelled with very pessimistic assumptions. As the FEM calculations by /Börgesson and Hernelind 1998/ show, these simple handbook calculations grossly overestimate the bending stresses in the canister insert. There are, however, no model simplification uncertainties of any importance for the safety assessment.

Effects of rock shear: There are no model simplification uncertainties of any major importance for the safety assessment.

Input data and data uncertainties

There are no uncertainties of any importance for the cases of isostatic loads and uneven loads during the saturation of the bentonite. There are, however, some uncertainties concerning the frequency and magnitude of rock shear movements following a de-glaciation. The consequences for the canister of a rock shear movement are sensitive to the bentonite density and there are some uncertainties concerning the possible variations in bentonite density in a deposition hole.

3.4.3 Deformation of copper canister from external pressure

Overview/general description

The copper canister is primarily a corrosion barrier. The mechanical strength of the copper canister is of subordinate importance, but the canister must withstand the loads associated with handling, transport and deposition. The copper must also possess sufficient ductility to allow straining, either plastically or by creep when the canister is deformed by the external load against the insert. Furthermore, the copper canister must sustain loads caused by deformations of the cast iron insert in response to external loads.

The following table shows how the process influences, and is influenced by, all canister variables.

When the load from the groundwater pressure and the swelling of the bentonite develops, the copper will deform plastically or by creep until it makes contact with the cast iron insert.

Boundary conditions

Swelling pressure from the bentonite from which even and uneven pressure situations can arise.

Flat (subhorizontal) rock displacements across the deposition hole can cause additional load on the canister.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	No Discussed in Section 3.4.6.		No	
Temperature	Yes The mechanical properties are temperature dependent.	Neglected The maximum mechanical load will occur after the period of increased temperature in the repository.	No	
Canister geometry	Yes The geometry and the choice of material will determine the strength of the copper shell.	The canister's mechanical stability is modelled using the actual canister dimensions.	Yes The copper will be deformed also at normal load.	Inherent in the model
Material composition	Yes The geometry and the choice of material will determine the strength of the copper shell.	The canister's mechanical stability is modelled using the actual canister materials properties.	No	
Mechanical stress	Yes Stresses are the direct cause of the deformation. High stresses in the material may also lower its strength.	Inherent in the model	Yes A deformation will alter the mechanical stresses.	Inherent in the model

Model studies/experimental studies

Calculations of stresses and strains in the copper canister after full pressure (assumed in the calculations to be 15 MPa) has built up in the repository have been performed by /Cakmak 1994/.

/Cakmak 1994/ presumed a rapid pressure build-up with the full hydrostatic pressure developed within an hour. The conclusions of the calculations were that:

- The copper shell is fully plasticized and contact occurs between the insert and the copper shell as a consequence of the external pressure. The lid is partially plasticized.
- The maximum strain of a few percent occurs locally on the inside of the shell near the transition to the lid and is caused by plastic deformation. The strain due to creep in the copper is negligible.
- The axial and tangential tensile stress lies around the yield stress and occurs, among other places, on the outside of the shell opposite the inner corner in the shell/lid transition and in the centre of the lid.

/Andersson et al. 2005/ have calculated the stresses and the strain in the copper shell for an outer pressure of up to 100 MPa and the additional strains after the shell is in contact with the insert are very small, less than 1%.

Handbook calculations have also been carried out for two cases where the swelling pressure is fully developed around the lower half of the canister, while the swelling pressure is 20 percent and 50 percent, respectively, around the upper half. The resulting upward force, caused by the differences in pressure against the end surfaces of the canister, is balanced by a shear force along the lower half of the copper canister. Calculations show that these load situations give rise to axial stresses in the copper wall equal to 8.4 and 21 MPa, respectively /Werme 1998/. These are low stresses compared with the yield stress of the copper material, which for annealed copper is about 45 MPa. For the copper canister, it can even be expected to be somewhat higher than that.

The response of the copper canister to a rock shear movement is discussed in Section 3.4.2.

Time perspective

The load from the groundwater pressure and the bentonite swelling pressure is expected to build up within a few years to a few decades. The maximum glacial load can be expected after tens of thousands of years.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

Isostatic load: The isostatic load on the canister over the one million year assessment period will be assessed in SR-Can. This load will cause the copper to deform until it is supported by the cast iron insert. This will result in a maximum strain of the copper of about a few percent and, therefore, is not expected to cause failure of the copper canister. Failure of the waste package will be controlled by the strength of the cast iron insert.

Uneven pressure build-up in the bentonite: The effects of this load situation are neglected, based on the calculations in /Werme 1998/ cited above.

Effects of rock shear: The likelihood of violating the criteria for rock shear movements that the canister can sustain (see Section 3.4.2) will be assessed based on estimated rock and buffer conditions. If violation occurs, the canister integrity will be assumed to be lost. Loss of integrity caused by creep relaxation of the stresses induced in the copper canister after a shear movement will be assessed based on the modelling by /Hernelind 2006/. When the creep strain exceeds the creep ductility of the copper material, the integrity will be lost.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

Isostatic load: The fundamental understanding is sufficient for the needs of the safety assessment.

Uneven pressure build-up in the bentonite: The fundamental understanding is sufficient for the needs of the safety assessment.

Effects of rock shear: The fundamental understanding of the response of the canister insert to a rock shear movement is sufficient for the needs of safety assessment.

Model simplification uncertainties

Isostatic load: There are no model simplification uncertainties of any importance for the safety assessment. The total maximum strain in the copper shell is shown to be well below both the tensile and creep ductility limits.

Uneven pressure build-up in the bentonite: The simplified model used in handbook calculations shows that the maximum stress in the copper shell is well below the yield stress of the copper.

Effects of rock shear: The materials models for the insert, the copper canister and the bentonite have no model simplification uncertainties of any importance for the safety assessment.

Input data and data uncertainties

There are no uncertainties of any importance for the cases of isostatic loads and uneven loads during the saturation of the bentonite. There are, however, uncertainties concerning the frequency and magnitude of rock shear movements following a de-glaciation. There are no data uncertainties of any importance in the modelling of the response of the canister to the rock shear movements.

3.4.4 Thermal expansion (both cast iron insert and copper canister)

Overview/general description

The canisters reach their maximum temperature (about 100°C) in the repository after about 10 years. The radial gap between the cast iron insert and the copper is 1–1.75 mm. If this gap is closed at this temperature due to creep or plastic deformation (this will cause a plastic strain in the copper of a few percent, see Section 3.5.3), cooling down to ambient temperature will cause a tensile strain in the copper shell due to the fact that the copper will shrink more than the cast iron when the temperature is decreasing. The resulting tensile strain can be estimated using the following formula:

$$\Delta\varepsilon = -\Delta T \cdot \Delta\alpha$$

where $\Delta\varepsilon$ is the increase of the tensile strain

ΔT is the temperature change of the system (10−100 = 90°C), and

$\Delta\alpha$ is the difference in thermal expansion coefficients for the copper shell and the cast iron insert ($17 \cdot 10^{-6} - 11 \cdot 10^{-6} = 6 \cdot 10^{-6}$ 1/°C)

$$\Delta\varepsilon = 0.00054\ (0.054\%)$$

This is of no importance when compared to the creep ductility of the copper (10%).

The following table shows how the process influences, and is influenced by, all canister variables.

Boundary conditions

The heat flow from the fuel and the pressure from the groundwater and swelling of the bentonite will form the boundaries for this process.

Model studies/experimental studies

See Overview/general description.

Time perspective

Heating to the maximum temperature takes place very quickly after deposition. Cooling to the ambient temperature takes hundreds to thousands of years.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is neglected based on the calculation results presented above.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of this process is sufficient for the needs of the safety assessment.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes It is caused by temperature.	Neglected based on simple calculations.	No	
Canister geometry	Yes The dimensions and the gap size control the final result.	Neglected based on simple calculations	No	
Material composition	Yes Determines the thermal expansion properties.	Neglected based on simple calculations	No	
Mechanical stress	No		Yes Pre-existing stresses may increase the stress level.	Neglected based on simple calculations

Model simplification uncertainties

Not relevant since the process is not further modelled in the safety assessment. (The model simplification uncertainties in the calculation used to justify the neglect of the process are insignificant.)

Input data and data uncertainties

Not relevant since the process is not further modelled in the safety assessment. (The data uncertainties in the calculation used to justify the neglect of the process are insignificant.)

3.4.5 Deformation from internal corrosion products

Overview/general description

If the copper canister is breached, water or water vapour will enter into the gap between the canister insert and the copper shell and give rise to anaerobic corrosion of the insert (see Section 3.5.1). If the water is also able to penetrate inside the lid on the insert, anaerobic corrosion of the inside of the channels for the fuel assemblies will occur.

In both cases, this leads to a build-up of corrosion products, which in turn causes mechanical stresses in the canister.

The sequence of events leading to the development of these stresses is expected to be proceeding as follows:

Water runs into the gap between the canister insert and the copper shell and into the canister insert, with anaerobic corrosion resulting. This leads to hydrogen gas generation, which increases the pressure inside the canister, whereupon the inflow of water decreases. This gas pressure is, however, limited to the opening pressure of the bentonite (see the SR-Can Buffer Process report, /SKB 2006g/) and will not be able to cause any deformation of the outer copper shell. After some time, the transport of water or water vapour into the canister by diffusion will be greater than the leakage of water into the canister due to the pressure difference. The time required for this varies with the size of the penetration in the copper canister and the corrosion rate, and is reasonably expected to be thousands of years. The inward diffusion of water vapour will prevent corrosion from ceasing entirely. The process leads to a slow build-up of corrosion products. These products occupy a larger volume than the equivalent quantity of cast iron, and with time an internal mechanical pressure is built up against the copper canister. This leads to local deformation and ultimately failure of the copper canister.

The following table shows how the process influences, and is influenced by, all canister variables.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved. The corrosion process is coupled to the inward transport of water for which boundary conditions are discussed in Section 2.3.1.

Model studies/experimental studies

Different evolutions of an initial canister defect with subsequent corrosion of the canister insert have been modelled by /Bond et al. 1997/.

Regardless of the size of the canister penetration and the corrosion rate, a sufficiently large hydrogen gas pressure is expected to have built up after some time to prevent liquid water from entering the canister. After that, the corrosion will nevertheless be able to continue due to the fact that water vapour can diffuse into the canister.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	No The temperature dependence of the corrosion rates is negligible.		No	
Canister geometry	Yes The canister geometry determines the size of the gap.	Obvious The actual geometries are considered.	Yes May cause expansion of the copper shell.	Several alternative scenarios that may lead to an enlargement of the initial failure are discussed
Material composition	Yes The material composition and the nature of the corrosion products determine the degree of volume expansion, which leads to deformation.	Obvious The actual materials are considered.	No	
Mechanical stress	No		Yes Stresses will build up in the canister materials.	Several alternative scenarios that may lead to an enlargement of the initial failure are discussed

/Bond et al. 1997/ developed a model for this diffusion-limited corrosion. The purpose was to predict how the solid corrosion products build up in the gap between insert and copper canister. The model takes into account the change in the size of the gap during the corrosion process, possible changes in the corrosion rate, and the escape of corrosion-generated hydrogen gas from the canister.

The gap is expected to eventually fill with magnetite, Fe_3O_4, and the calculations were used to analyze the consequences of this for the strength of the canister. Two situations were analyzed: one extreme case where the entire outside surface area of the insert corroded and one case where the corrosion was limited to an area around the original defect. The latter case progresses into the former after the copper material around the original defect begins to give way. This is calculated in the model to take around 20,000 years. According to the calculations, from the time of the initial penetration, it takes at least 100,000 years before more extensive damage~~s~~ occurs on the copper canister.

Experimental studies of the possible pressure build-up from the corrosion products showed no jacking effects caused by expanding corrosion products /Smart et al. 2003/. Further studies by /Smart et al. 2006/ gave similar results.

Time perspective

The process will start immediately after penetration of the outer copper shell.

Natural analogues/observations in nature

Some archaeological analogues are discussed by /Smart et al. 2003/.

Handling in the safety assessment SR-Can

For an intact canister, the process is neglected.

In the case of a canister failure, the process is treated in the SR-Can main report as a part of an integral description of the evolution of the canister interior after damage (see further Section 2.3.1).

Due to the uncertainties in the development of the annulus between the copper and the cast iron, several alternative cases are considered. As a base case, the corrosion in the filled annulus ceases and the insert remains tight. Conceivable alternative cases are:

- Corrosion in the filled annulus continues and the copper shell expands followed by creep relaxation of the bentonite.
- Corrosion in the filled annulus continues and the copper shell expands followed by compression of the bentonite.
- Corrosion in the filled annulus ceases but continues inside the cavity of the insert.

All these evolutions will eventually lead to failure of the cast iron insert and an enlargement of the initially small penetration in the copper shell. This will allow an increase in the release rate of radionuclides from the canister.

The consequences of all these developments can be pessimistically bounded by estimating conservative values of the time at which the larger failure occurs and of the size of this failure. This is further developed in the SR-Can main report.

Boundary conditions: The inward transport of water is included in the study by /Bond et al. 1997/.

Handling of variables influencing this process: The influencing variables in the table are included in the modelling by /Bond et al. 1997/ and in the cases considered in the integrated treatment of the process.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There are uncertainties surrounding the behaviour of the canister insert under the loading caused by growing corrosion products and over the available surface area as corrosion progresses. The experimental studies /Smart et al. 2003, 2006/ seem to indicate that the corrosion products do not exert enough pressure to displace the copper shell. Further studies will, however, be necessary and have been initiated at the Äspö Hard Rock Laboratory.

As discussed above, despite these uncertainties, it is possible to put an upper bound on the consequences of this process in the safety assessment.

Model simplification uncertainties

Modelling requires assumptions regarding the properties of the corrosion products. These, and other aspects of the model, are not verified experimentally.

Rather than attempting a justification of model simplifications, the consequences of this process are given a pessimistic upper bound in the safety assessment, as discussed above.

Input data and data uncertainties

The corrosion rates of freely exposed metal are relatively well known. There are, however, large uncertainties concerning the form and distribution of the corrosion over the insert surface area when the metal is exposed to a limited amount of water or water vapour. There is also uncertainty over the density and porosity of corrosion products and, therefore, the degree of expansion.

Rather than seeking defensible treatment of data uncertainties, the consequences of this process are given a pessimistic upper bound in the safety assessment as discussed above.

3.4.6 Radiation effects

Overview/general description

Neutron and gamma-radiation from the fuel can give rise to minor material changes in the cast iron insert and the copper canister.

The effects of irradiation of the canister materials over a long period of time have been discussed by /Guinan 2001/. By comparing with experiments in the literature, /Guinan 2001/ concluded that the magnitude of any physical property changes (e.g. yield stress, creep rates, enhanced solute segregation, dimensional changes, or brittleness) resulting from exposure to neutron and gamma radiation over the one million year time period of the safety assessment will be negligible.

The following table shows how the process influences, and is influenced by, all canister variables.

Neutron irradiation might also lead to the formation of activation products in the canister insert.

Boundary conditions

There are no relevant boundary conditions for this process other than the physical boundary set by the geometries of the components involved. (Neutron and gamma- radiation from the fuel is required for this process to occur.)

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	Yes It causes the process	Neglected Based on modelling results.	No	
Temperature	Yes At repository temperatures any effect can be neglected.	Neglected Based on modelling results.	Yes The heating caused by the process is negligible	Neglected Based on modelling results.
Canister geometry	No		No	
Material composition	No		Yes Radiation embrittlement will occur to some extent in all materials.	Neglected Based on modelling results.
Mechanical stress	No		No	

Model studies/experimental studies

See Overview/general description.

Time perspective

The neutrons come from spontaneous fission and from (α, n) reactions in the fuel. Both processes diminish greatly with time, and 1,000 years after deposition the radiation dose has declined by a factor of thirty /Håkansson 1999/.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is neglected based on the results presented in /Guinan 2001/ and cited above.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

The mechanistic understanding of this process is deemed sufficient for the needs of the safety assessment.

Model simplification uncertainties

Not relevant since the process is not further modelled in the safety assessment. There are no relevant model simplification uncertainties of any importance used in /Guinan 2001/ to justify the neglect of the process.

Input data and data uncertainties

Not relevant since the process is not further modelled in the safety assessment. There are no relevant data uncertainties of any importance used in /Guinan 2001/ to justify the neglect of the process.

3.5 Chemical processes

3.5.1 Corrosion of cast iron insert

Overview/general description

Water in the canister cavity and in the annulus between copper and cast iron will corrode the cast iron insert. As a consequence of this process, the cast iron insert's integrity and mechanical strength may be jeopardised. Another result is the formation of gaseous hydrogen and solid corrosion products, of which the latter can exert pressure against the copper canister.

The process is central to the canister's hydraulic, mechanical and chemical evolution if the copper shell should be breached.

Corrosion can be caused by water inside an intact canister. This possibility cannot be ruled out since some water may be present in the canister enclosed in fuel pins. Once oxygen inside the canister has been consumed by corrosion, this water will cause anaerobic corrosion on the insides of the insert.

Corrosion can also be caused by water that has entered a defective canister. Groundwater at repository depth is expected to be oxygen-free. The cast iron insert will, therefore, corrode anaerobically to generate hydrogen gas and form a solid corrosion product, most likely magnetite, and also some dissolved Fe(II). The magnetite is expected to consist of a thin adherent epitaxial layer and an outer, looser layer with poor adhesion. The resistance of the inner layer to ion transport will determine the corrosion rate. Since this layer appears to achieve a constant thickness while the outer non-protective layer continues to thicken as corrosion continues, the corrosion rate is expected to remain constant over a long period, see below. Alternatively, if the water supply is low, the corrosion rate will be determined by the availability of water.

The following table shows how the process influences, and is influenced by, all canister variables.

The corrosion rate of iron under anoxic conditions is dependent on the water composition and the temperature. The corrosion products and the corrosion process affect the chemical conditions in the cavity in the canister mainly by generating high concentrations of dissolved hydrogen gas and small concentrations of dissolved Fe(II). The corrosion rate has been shown to be independent of both the hydrogen gas pressure and the concentration of Fe^{2+} in the system /Smart et al. 2002ab/. This suggests that the corrosion rate is most likely determined transport processes in the magnetite layer on the iron surface (see below).

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	Yes Gamma-radiation may influence the corrosion that takes place during the first several hundred years.	See 3.5.3	No	
Temperature	Yes The influence is, however, of marginal importance.	Neglected	No	
Canister geometry	Yes Of marginal importance.	Neglected	Yes The corrosion products have a larger volume than the metal.	Neglected After failure the influence of the corrosion products on the canister geometry and on transport resistance for radionuclides is neglected.
Material composition	Yes Different materials have different corrosion properties.	Actual insert material is considered in the evaluation.	Yes Corrosion products are formed.	Actual corrosion products are considered in the evaluation.
Mechanical stress	Yes Possible environmentally assisted cracking.	See 3.5.3	Yes The corrosion products may exert a pressure on the canister components.	Neglected After failure the influence of the corrosion products on the stress levels in the insert is neglected.

Boundary conditions

The boundary conditions for this process are the physical boundary set by the geometries of the components involved and the composition of water in the canister void. The corrosion process is also coupled to the inward transport of water for which boundary conditions are discussed in Section 2.3.1.

Model studies/experimental studies

Corrosion caused by water inside an intact canister: The fuel elements will be dried before being transferred to the canister, but some water may nevertheless be transferred. A very pessimistic assumption would be that one fuel pin per fuel element with maximum water fill is transferred to each canister.

The scope of the corrosion this could cause can be modelled using a simple mass balance approach:

The total quantity of water that can be accommodated in the void in a fuel pin is 50 g. It is expected that no more than 12 fuel pins containing that amount of water will be transferred to the canister. The total maximum amount of water will, therefore, be 600 g. If this water reacts solely with the iron in the canister and is assumed to form Fe_3O_4, a maximum of about 1,400 g of iron could corrode**.** This will result in a relatively uniform corrosion depth of less than 10 μm for the cast insert.

Corrosion caused by water that has entered a defective canister: Anaerobic corrosion of cast iron has been studied experimentally by /Smart et al. 2002ab/, who found that the corrosion rate is well below 1 μm/y after a few thousand hours, even in the most aggressive water tested (Äspö groundwater KAS-03), see Table 3-2.

In the reaction between cast iron and water, magnetite and hydrogen are produced. Initially, the corrosion rate is quite high but falls off rapidly to very low rates as the surface film of magnetite develops. The rate of hydrogen production at the highest measured long-term corrosion rate is about 0.5 $dm^3/(m^2 \cdot y)$ /Smart et al. 2002b/. The total internal surface area in the canister insert is approximately 33 m^2, which means the annual production of hydrogen would be 16 dm^3 at normal pressure or 0.33 dm^3 at the pressure at repository level (500 m depth).

The corrosion rate has proved to be independent of both the hydrogen gas pressure /Smart et al. 2002a/ and the concentration of Fe^{2+} /Smart et al. 2002ab/ in the system. The long-term corrosion rate is also independent of whether or not the water is present as a liquid or as vapour /Smart et al. 2002b/. This suggests that the corrosion rate is most likely determined by the ion transport properties of the magnetite layer on the iron surface:

The magnetite consists of two layers: a thin, strongly adherent layer and an outer, looser layer with poor adhesion /Smart et al. 2002b/. Smart et al. explain the formation of these two layers by saying that the adherent layer is formed directly on the metal surface by the reaction:

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2$$

while the looser layer is probably formed by the precipitation of dissolved iron:

$$Fe + 2H_2O \rightarrow Fe^{2+} + H_2 + 2OH^-$$

$$3\,Fe^{2+} + 6OH^- \rightarrow Fe_3O_4 + 3H_2 + 2H_2O \qquad \text{(the Schikkor reaction)}$$

Table 3-2. Composition (ppm) of synthetic Äspö groundwater KAS-03.

Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	HCO_3^-	SO_4^{2-}	pH
3,000	7	4,400	50	12,000	11	710	7–8

The adherent layer forms very quickly and then does not increase further in thickness. Continuing corrosion leads to the thickening of the looser non-protective layer. Corrosion is controlled by ion transport across the adherent layer and should continue at a constant rate over long periods of time. The adherent layer forms very quickly and then does not increase further in thickness. Subsequently, corrosion would be supported by ion transport across this layer.

The corroding iron insert will influence the water chemistry in the void in the canister by generating high concentrations of dissolved hydrogen gas and small concentrations of dissolved Fe(II). Calculations with EQ3/6 show that the solubility of dissolved iron in the void in the canister is, as expected, dependent on the redox conditions, but can scarcely be higher than 10^{-7} mole/dm^3 (Calculations performed at SKB for this report). The hydrogen will be dissolved to a concentration equivalent to a maximum hydrogen gas pressure of max. 18 MPa. The iron concentrations are too low to be of any crucial importance for the water chemistry. The hydrogen will have no influence on the corrosion of the iron insert (as mentioned above).

Time perspective

At a corrosion rate of 1 μm/y, metallic iron will be present for tens of thousands of years after failure of the copper canister. The length of time iron is present is inversely proportional to the constant corrosion rate.

Natural analogues/observations in nature

There are some studies of archaeological artefacts reported in the literature. Recent studies by /Neff et al. 2003, 2005/ review some newer studies as well as data from their own investigations. /Neff et al. 2005/ carefully studied the corrosion products on artefacts from the 2nd century AD to the 16th century AD. In a second publication, /Neff et al. 2006/ analyse the archaeological artefacts in order to determine the average corrosion rate of low carbon steel after long burial periods. The corrosion products found indicate some of the corrosion attack took place with oxygen present. Nevertheless, the colclusion from the study is that the estimated average corrosion rates do not exceed 4 μm per year.

In /Neff et al. 2003/, the authors review some recent studies as well as presenting data of their own. The measured or estimated corrosion rates vary over a relatively large range and the corrosion products, when identified, indicate that oxygen had been present in the system. The reported rates range from 0.01 μm per year to extreme values of 1,000 μm per year. In one site where the conditions have been oxygen free and the carbonate content in the water has been high (10^{-2} M), siderite is identified as the main corrosion product /Matthiesen et al. 2003, 2004/. This site at Nydam Mose in Denmark was waterlogged and the buried objects are military equipment sacrificed in the period 200–500 AD. The estimated corrosion rates range from an upper limit of 5 μm per year down to 0.03 μm per year or less. The average corrosion rate for 151 analysed lances is about 0.2 μm per year.

Handling in the safety assessment SR-Can

Even with a pessimistic assumption of 600 g water per canister, the effects of the corrosion of the canister insert prior to failure of the copper canister can be neglected.

In the case of a canister breach, the process is treated in the SR-Can main report as part of an integral description of the evolution of the canister interior after damage (see further Section 2.3.1).

The treatment aims at bounding the consequences of the effects of the set of integrated processes. This is further developed in the SR-Can main report.

Boundary conditions: The boundary conditions mentioned above are included in the integrated treatment, see Section 2.3.1.

Handling of variables influencing this process: Of the influencing variables in the table, the canister geometry and the material properties are included in the integrated treatment, see Section 2.3.1. The temperature is though assumed to be constant and equal to about 15°C and anaerobic conditions are assumed to prevail in the canister interior. Radiation effects are neglected based on the discussion in Section 2.5.2.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There are no major uncertainties in the understanding of the corrosion of iron under oxygen free, reducing conditions.

Model simplification uncertainties

In a breached canister, the process is modelled as a constant corrosion rate unless the supply of water is limiting. Several developments, in which this process is integrated with a number of others, are considered in order to bracket the possible scenarios.

Rather than seeking justification of model simplifications, the consequences of this process are given a pessimistic upper bound in the safety assessment as discussed above.

Input data and data uncertainties

Intact canister: There are great uncertainties surrounding the quantity of water that could conceivably be introduced into a canister by mistake. The great majority of canisters will probably not contain any water. The uncertainties here are of no importance, since the extent of corrosion pessimistically achievable would be negligible.

Damaged canister: The uncertainties regarding the corrosion rate are discussed in the SR-Can Data Report /SKB 2006b/. These uncertainties are relatively small, and the corrosion rate can be expected to be less than 1 μm/y, with 0.1 μm/y being a likely value. The experimental investigations show that if the protective corrosion product layer of magnetite is damaged, it will reform very quickly and the measured corrosion rates can be assumed to apply even for long periods of time.

3.5.2 Galvanic corrosion

Overview/general description

If the copper shell is breached and groundwater comes into contact with the cast iron insert, electrochemical reactions on the copper surface will influence the corrosion of the insert. The consequences of galvanic corrosion have been investigated first by /Blackwood and Naish 1994/ and recently by /Smart et al. 2005/.

If the groundwater contained oxygen, the rate of cast iron corrosion was very high, up to 100 μm per year /Smart et al. 2005/. At the time, however, of water intrusion into the canister the water is expected to be oxygen-free and reducing. In oxygen-free water, the measured corrosion rates of cast iron galvanically coupled to copper were in the same range as those measured for cast iron in the absence of galvanic coupling to copper. /Smart et al. 2005/.

The following table shows how the process influences, and influenced by, all canister variables.

Boundary conditions

The boundary conditions for this process are the physical boundary set by the geometries of the components involved and the composition of the water in the canister void. The corrosion process is coupled to the inward transport of water for which boundary conditions are discussed in Section 2.3.1.

Variable	Variable influence on process Influence present?	 Handling of influence	Process influence on variable Influence present?	 Handling of influence
Radiation intensity	No		No	
Temperature	Yes Corrosion reactions are generally temperature dependent.	Neglected The influence of galvanic corrosion under oxygen-free, reducing conditions lies within the margins of error for the corrosion rate of the iron insert.	No	
Canister geometry	No There will always be a galvanic contact regardless of geometry.		No	
Material composition	Yes Galvanic corrosion requires two metals.	Neglected The influence of galvanic corrosion under oxygen-free, reducing conditions lies within the margins of error for the corrosion rate of the iron insert.	No	
Mechanical stress	No		Yes Growth of corrosion products will cause stresses in the canister materials.	Neglected The influence of galvanic corrosion under oxygen-free, reducing conditions lies within the margins of error for the corrosion rate of the iron insert.

Model studies/experimental studies

See Overview/general description.

Time perspective

The process takes place after canister penetration.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The influence of galvanic corrosion under oxygen-free, reducing conditions lies within the margins of error for the corrosion rate of the iron insert and is, therefore, not dealt with specifically.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There are no major uncertainties in the understanding of galvanic corrosion.

Model simplification uncertainties

In oxygen-free water, the effects will be small and the uncertainties surrounding the exact contribution made by galvanic corrosion are judged to be negligible compared with the uncertainties in assessments of the corrosion rate. A cautious hypothesis for the corrosion rate for anaerobic corrosion of the canister insert will cover any contributions from galvanic corrosion.

Input data and data uncertainties

The uncertainties are negligible compared to those handled in Section 3.5.1

3.5.3 Stress corrosion cracking of cast iron insert

Overview/general description

The stress corrosion cracking (SCC) of metals requires a combination of static tensile stresses, a special chemical environment and, a susceptibility of the material. Unfavourable combinations of these conditions can lead to the initiation and propagation of cracks. The propagation rate can vary over a wide range, from 10^{-9} mm/s to 10^{-1} mm/s.

In a canister with an intact copper shell, nitric acid from the radiolysis of residual quantities of air could conceivably cause SCC in areas of tensile stress~~es~~ in the cast iron insert /Blackwood et al. 1995/.

Shortly after deposition and closure, the temperature of the insert nearest the fuel is expected to be over 150°C. At this temperature there will be no water in liquid form in the canister, and the relative humidity is too low for a water film to form on the metal surface, even if there was water in the canister trapped in the fuel rods. (The largest quantity expected from this source is estimated to be 600 g, equivalent to the voids in 12 fuel pins.) For a water film to form, a relative humidity of approximately 40 percent is required. Under these conditions, nitric acid is not stable but decomposes to NO_2, the dominant radiolysis product in dry air /Reed and Van Konynenburg 1991ab/. For SCC conditions to develop the water and oxygen content of the canister must survive until the temperature falls sufficiently to allow water/nitric acid condensation on the canister; i.e. their consumption by prior corrosion must be extremely low.

For SCC to occur, a corrosive environment must exist while tensile stresses are present in in the material. In the deep repository, the canister insert is under external pressure and tensile stresses occur on the cast insert only locally and in small areas, according to calculations /Dillström 2005/. It is therefore highly improbable that SCC could lead to penetrating cracks in the canister, and above all that this would jeopardise the integrity of the canister. The risk for SCC caused by nitrate can be lowered by replacing the air in the canister with a noble gas.

The following table shows how the process influences, and is influenced by, all canister variables.

Stress corrosion cracking (SCC) of metals can occur under a combination of static tensile stresses, a special chemical environment and, metallurgical susceptibility. Unfavourable combinations of these conditions can lead to the formation and propagation of cracks. The temperature dependence of the extent of this process for carbon steel in nitrate follows a typical Arrhenius behaviour /Beavers et al. 1985/.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	Yes The agents that may cause SCC are produced through radiolysis.	Neglected Process neglected since only tensile stresses only occur locally in small areas.	No	
Temperature	Yes Negligible influence on this process.	Neglected	No	
Canister geometry	No		No	
Material composition	Yes Different materials have different sensitivities to SCC.	Actual insert material is considered.	No	
Mechanical stress	Yes Tensile stresses required. Compressive stresses prevent SCC.	Neglected Process neglected since only tensile stresses only occur locally in small areas.	Yes SCC relaxes tensile mechanical stresses.	

Boundary conditions

Nitric acid from radiolysis of residual quantities of moist air in the void inside the canister (the fuel boundary) could conceivably cause SCC in areas with tensile stresses in the cast iron insert.

Model studies/experimental studies

See Overview/general description.

Time perspective

It is estimated that an equilibrium concentration of nitric acid will have been reached a few decades after closure of the canister.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

Stress corrosion cracking, if it occurs, is deemed to be of no importance for the integrity of the canister.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There are uncertainties in the understanding of stress corrosion cracking, but they will be of no importance for the safety analysis, since the extent of the process will be limited.

Model simplification uncertainties

Stress corrosion cracking will not be modelled.

Input data and data uncertainties

The probability of SCC jeopardising the canister integrity is deemed to be small, since only local areas in the insert have tensile stresses. The greatest uncertainty concerns the quantity of water that may have been introduced into the canister.

3.5.4 Corrosion of copper canister

Overview/general description

A range of studies over several decades (see e.g. /King et al. 2001/ for a review) have identified the following substances as capable of corroding the copper canister material under repository conditions:

- oxygen introduced during the repository operating period
- oxygen trapped in the air-filled pore spaces in the buffer or available in the groundwater transported through the buffer
- nitric acid formed by gamma-radiolysis of nitrogen compounds in moist air in the gap between canister and buffer
- sulphide brought in from the buffer or from the groundwater via the buffer

The corrosion processes are marginally affected by the changes in temperature expected in the deep repository. The consequences of corrosion are corrosion products and a change in the thickness of the copper shell. After very long time spans the consequence is a breach of the shell's isolation.

The copper shell is the canister's corrosion barrier. Its design purpose is to provide protection against penetration of the canister by corrosion in the repository for a long time after water saturation. It is also supposed to provide protection against penetration of the canister by atmospheric corrosion before deposition and after deposition before water saturation.

After water saturation, oxygen will still be present in the canister's surroundings. This oxygen is consumed by reactions with the copper canister and by reactions with minerals in the bentonite buffer or the host rock. Microbes in the deposition tunnel backfill may also play an important role in oxygen consumption. /Wersin et al. 1994a/ estimated the time this takes to be between 10 and 300 years. Recent numerical calculations /Grandia et al. 2006/ coupling chemical processes consuming oxygen with the hydrodynamic saturation of the backfill have been used to estimate the time scale for reaching anoxic conditions in the tunnels of the repository. The study shows that several inorganic O_2 consumption processes may take place with the accessory minerals present in the bentonite in the buffer and in the backfill. As in the study by /Wersin et al. 1994a/, the calculated oxygen consumption times are highly dependent on the postulated value for the surface area of the reacting minerals. Nevertheless, the study concludes that anoxic conditions are reached after a period of the order of one month after the backfill becomes water saturated.

The calculations do not include the possible effects from microbial respiration, although the density of the backfill is low enough to allow microbial activities. The effect of microbes would be to shorten the time to reach anoxic conditions in the backfill.

When the oxygen is consumed, the reducing conditions that prevailed before the repository was excavated are expected to be restored. During the period when oxygen is present, one might argue that localized corrosion in the form of pitting is also possible, which can lead to localized deeper corrosion attacks than the averagedue to general corrosion. The scope of pitting is often described by the pitting factor, which is the ratio between the greatest corrosion depth achieved and the depth of the general corrosion process. As will be argued below, however, pitting corrosion under repository conditions will be very unlikely.

When the water is reducing, copper should be immune to corrosion, see e.g. /King et al. 2001/. However, under deep repository conditions, dissolved sulphide or very high chloride concentrations are conceivable. High chloride concentration in combination with very low pH could cause copper corrosion in oxygen free water. However, since deep groundwaters are neutral or slightly alkaline and the buffer capacity of the bentonite and the rock counteracts acidification, dissolved sulphides are, in practice, the only corrosive substances that can react with the copper canister after the oxygen in the repository has been consumed. At the proposed repository depth the groundwaters have very low sulphide concentrations, much lower than $5 \cdot 10^{-5}$ mol/dm^3, and the solubility of the sulphide minerals present in the bentonite is at most of the same order of magnitude. This means that the corrosion of the copper canister due to sulphides will be controlled by the availability and supply of sulphides from the groundwater and the buffer.

A review of copper corrosion under expected repository conditions has been performed by /King et al. 2001/. In that review the corrosion processes and mechanisms discussed below are elaborated in more detail.

Copper corrosion of initially entrapped oxygen

In the presence of oxygen, copper will be oxidized to Cu_2O or CuO in pure water, depending on the redox potential. In the presence of ground water anions, hydroxysulphates, hydroxycarbonates or hydroxychlorides can form. These compounds can also form under atmospheric corrosion conditions.

The total extent of corrosion under aerobic conditions will be limited by the available amount of oxygen trapped in the repository after closure. This amount can be calculated to be 560 moles per canister corresponding to a maximum depth of corrosion of 840 μm if it were evenly distributed over the canister surface. Most of the residual oxygen is present in the tunnel backfill and is expected to be consumed not only by the copper corrosion reaction but also through reactions with oxidisable minerals and through microbial activities (see above). A realistic estimate would be that only a few percent of the oxygen will be consumed through corrosion reactions with copper, i.e. the corrosion depth would be 10 micrometres or less.

Corrosion under the expected conditions in the deep repository after water saturation has been well studied /Swedish Corrosion Institute 1978, 1983, Werme et al. 1992, Wersin et al. 1994b, King et al. 2001/.

1. /King et al. 2001/ discuss the results of a number of corrosion studies that have been performed under conditions that simulate the canister near-field environment. Copper coupons have been exposed to compacted buffer wetted by (initially) aerated saline pore water for extended periods of time (up to 2 years) usually at elevated temperature /Aaltonen and Varis 1993, Karnland et al. 2000, King et al. 1992, Litke et al. 1992/. These experiments simulate the likely environmental conditions soon after emplacement of the canisters and saturation of the buffer material. /King et al. 2001/ concluded that despite the relative aggressiveness of the conditions in these tests, no evidence for pitting is observed. Thus, both /Aaltonen and Varis 1993/ and /Karnland et al. 2000/ report no localized corrosion of Cu exposed to compacted clay over periods of up to 2 years. The only instance of non-uniform corrosion reported under such conditions is a so-called under-deposit corrosion reported by /Litke et al. 1992/. In this case the whole surface had been corroded resulting in an uneven general corrosion with variations on corrosion depth of 30 μm for an average corrosion depth of slightly over 40 μm. These data indicate that no pitting is to be expected, but that there may be unevenness around the average corrosion depth of roughly ± 30 to 50 μm.

2. Another approach to predicting the extent of localized corrosion on Cu canisters is to make projections based on observed pit depths. Because pitting has not been observed on Cu exposed to simulated repository conditions, the required pit depth data has been taken from literature studies of the long-term burial of Cu alloys /Romanoff 1989/ and from an analysis of pit depths on three lightning conductor plates /Hallberg et al. 1984/ and on archaeological artifacts /Bresle et al. 1983/. Whilst the environmental conditions and Cu alloys are different from those to be used in a repository, these studies have the great advantage of having been "conducted" over long periods of time. Thus, the long-term soil corrosivity measurements of /Romanoff 1989/ were conducted for times up to 14 years. The lightning conductor plates studied by /Hallberg et al. 1984/ and been buried for 50 to 80 years. The Bronze Age artefacts studied by /Bresle et al. 1983/ had been exposed for an estimated period of 3,000 years. These pit depths can be used to estimate the pitting factor (PF), defined above, and for which PF = 1 for general corrosion. From these studies, /Romanoff 1989/ reported the highest pitting factor of 25, but observed that it decreased with time. The results of /Bresle et al. 1983/ indicated a probable pitting factor of 2 to 5. /Hallberg et al. 1984/ found a pitting factor of 5 for two of the lightning conductors and no pitting for the third. These data indicate that, for extended exposure times, a pessimistic estimate of the pitting factor would be 5. The use of a pitting factor, however, will overestimate the pit depth at larger general corrosion depths, since general corrosion destroys the separation of anodes and cathodes required to drive deep pitting.
3. An alternative use of the same pit depth data has been developed in the Canadian programme /King and Kolár 2000, King and LeNeveu 1992/. The data were analysed using extreme-value statistics, in which the deepest pits on a collection of samples (or within a given area of the surface of one or more samples) of the same exposure time are fitted to an extreme-value distribution of the form,

$$F(x) = \exp[-\exp(-ax + b)]$$

where F(x) is the cumulative probability that the depth of the deepest pit is less than or equal to a depth x, a is the scale (or shape) parameter, and b/a is the location parameter of the distribution. The fitting procedure produces vales for a and b for a given set of data for a given exposure period. Fitting several sets of data with different exposure periods gives the time dependence of the scale and location parameters. From these fits and calculations the time dependence of the cumulative probability (F(x)) can be calculated. Then the probability of a pit of a certain depth can be calculated for the full population of canisters emplaced in the repository. Applying these data to Swedish canisters, one can calculate that there is a probability of 10^{-6} of a pit exceeding about 7.5 mm after 10^6 years. This approach predicts, however, very deep pits after shorter exposure times. With the same probability (10^{-6}) it predicts a pit 5 mm deep after 10 years. This is a result of the very high pitting factors observed on some specimens by /Romanoff 1989/.

Based on the experimental data available the most realistic understanding of the pitting during the period when oxygen is present is that it will have the appearance of uneven general corrosion as discussed under point 1 above. Those data were obtained under conditions more closely simulating actual disposal conditions, whereas some of the data discussed in the statistical avaluations (2 and 3 /Bresle et al. 1983/) were obtained after exposure in poorly characterized environments. However, they clearly illustrate the tendency of pit growth rates to decrease with exposure time.

After consumption of the initially present oxygen, reducing conditions are expected but the possibility of oxygen intrusion for altered geochemical conditions exists and can be handled by mass balance calculations if the amount of intrusive oxygen can be estimated.

Corrosion caused by nitric acid during the buffer saturation phase: Gamma irradiation of moist air in the canister-buffer gap leads to the formation of nitrogen oxides, which in contact with water form nitric acid. /Marsh 1990/ shows a simple method to make a rough calculation of the quantity of nitric acid produced. The rate of formation of nitric acid is given by:

$$d[HNO_3]/dt = G \cdot V \cdot \rho \cdot D_0/A_v \cdot e^{-0.693\, t/T}$$

where G is the G value (in number of molecules/eV), V the irradiated air volume (dm^3), ρ the density of the air (g/dm^3), D_o the initial dose rate (eV/g·y), A_v Avogadro's number, t the time (years) and T the half-life (years) of the radiation source. If it is assumed that the gamma radiation source has a half-life of 30 years, G = 0.02 molecules/eV and an air gap of 5 mm exists around the canister (V = 85 dm^3), then a total production of 0.008 mol of nitric acid is obtained. This is a small quantity, which will have a negligible affect the life of the canister.

After water saturation, radiolysis of water near the canister will occur. This will lead to the formation of oxidants and hydrogen. If these radiolysis products are not removed, equilibrium will soon be reached whereby the further production of hydrogen and oxidants ceases. If the radiolysis products are removed, however, for example by the diffusion of hydrogen away from the canister or by reaction of the oxidants with copper or other oxidisable species in the system, radiolysis can continue. Calculations of radiolysis outside a canister have been made by /Christensen and Pettersson 1997/. An estimate of the maximum possible amount of oxidised copper can be made if one assumes that the oxidation of copper will be as efficient as the oxidation of dissolved Fe(II) in Christensen's calculations. Assuming that the oxidants present in a 5 mm water layer surrounding the canister reach and react with the copper surface, a maximum of 3.2 kg of copper can be oxidised over 300 years. This would translate to a corrosion depth of about 3 µm. However, the experimental observations of /King et al. 2001/ indicate that the overall effect of radiation on copper corrosion under oxidising conditions will be negligible or even result in a lowering of the corrosion rate.

Corrosion in the absence of oxygen

In the absence of oxygen, copper is thermodynamically immune to corrosion in pure water. Corrosion with hydrogen gas generation can take place if the water contains dissolved sulphides. The corrosion products are then Cu_2S or CuS. High chloride concentrations in combination with low pH values can also cause corrosion on copper. In the former case, the corrosion rate will be controlled by the inward transport of sulphide ions to the canister surface, and in the latter case by the outward transport of copper chloride complexes.

After the oxygen has been consumed and reducing conditions have been reinstated, copper corrosion under deep repository conditions will be controlled by the availability of dissolved sulphides. This sulphide may be present in the bentonite buffer and backfill and also in the deep groundwaters. The concentrations of dissolved sulphide in the near field are controlled by the solubility of sulphide minerals in the bentonite, and in the rock the concentrations are generally well below $5 \cdot 10^{-5}$ mol/dm^3. At these low concentrations, the transport of sulphide from or through the buffer to the copper surface is very slow and this transport resistance will completely control the rate of the corrosion process.

/King et al. 2001/ make an estimate of the extent of the corrosion attack over 1 million years and also compare their results with previous estimates /Wersin et al. 1994b, Werme et al. 1992, Swedish Corrosion Institute 1983, Johnson et al. 1996/. All estimates result in corrosion depths of no more than a few millimetres in one million years.

Copper corrosion after water saturation; bacterial corrosion: Normally, bacterial reduction of sulphate to sulphide does not pose any threat to the integrity of the canister, since this does not lead to elevated levels of dissolved sulphide in the groundwater. The worst case would be if the bacteria formed a biofilm on the canister surface or were present in the buffer near the canister. Corrosion would then be controlled by the transport of sulphate to the canister, and could be considerably accelerated, since the transport of sulphate is expected to be much faster than the transport of sulphide, due to sulphate concentrations in the bentonite of up to tens of mmol/dm^3.

Studies of the conditions for the growth of sulphate-reducing bacteria in compacted bentonite have been conducted since 1995 /Pedersen et al. 1995, 2000, Motamedi et al. 1996, Masurat and Pedersen 2004, Masurat, 2006/. The results showed that sulphate-reducing bacteria cannot survive in bentonite if the bentonite is compacted to a density corresponding to a swelling pressure of 2 MPa. /Masurat 2006/ also studied the microbial sulphate production as a function of swelling pressure. At 0.14 MPa bentonite swelling pressure and for the most favourable conditions for microbial growth, the measured the maximum microbial sulphide production rate, expressed as Cu_xS will be less than 1.5 pmol·mm^{-2}·day^{-1}. This corresponds to a corrosion depth of 4 μm in 1,000 years.

Sulphate corrosion: Inorganic sulphate corrosion of copper has been deemed impossible under repository conditions, since the reaction is kinetically inhibited /Swedish Corrosion Institute 1983, Grauer 1991/.

The following table shows how the process influences, and is influenced by, all canister variables.

Variable	**Variable influence on process**		**Process influence on variable**	
	Influence present?	**Handling of influence**	**Influence present?**	**Handling of influence**
Radiation intensity	No Radiation effects are negligible.		No	
Temperature	Yes Complex dependence on temperature depending on the relative importance of kinetic and transport steps	Neglected The corrosion is modelled using transport control and mass balance	No	
Canister geometry	No		Yes The canister dimensions will change due to corrosion, but the changes will be negligible.	Neglected
Material composition	Yes Different materials have different corrosion behaviour.	Actual copper material is considered.	Yes Corrosion products are formed.	Actual corrosion products formed under the foreseen conditions are considered.
Mechanical stress	Yes Dealt with in Section 3.5.5		No The growth of corrosion products is too slow to create additional pressure on the canister.	

The temperature in the repository will be elevated during the oxidizing phase, with a maximum temperature of up to 100°C on the copper surface. The corrosion reactions are temperature dependent, generally with increasing rates at higher temperatures. This is, however, of subordinate importance, since the diffusion of reactants is rate controlling and the diffusivity is much less affected by the temperature. The influence of the temperature on the chemical equilibria for the corrosion reactions is completely negligible for the temperature range encountered in the repository.

During a deglaciation there may be a possibility of intrusion of oxic water to the repository. As for sulphide corrosion, the transport of oxygen to the canister surface will be rate limiting. /King and Kolar 1997/ have modelled the effects of the presence of aerated groundwater at the repository/geosphere interface. It was found that there was sufficient Fe(II) in Canadian Shield granite to consume all O_2 in the groundwater for a period in excess of 1 million years. Therefore, the rock could be expected to act as both a mass transport and as a redox barrier if aerated water was introduced at repository depth during a deglaciation period.

Boundary conditions

The boundary condition for the copper corrosion process will be the rate of supply of corrosive agents from the near field to the canister – buffer boundary.

Model studies/experimental studies

See Overview/general description.

Time perspective

Sulphide corrosion will proceed as long as metallic copper is available and sulphide is present in the buffer, the deposition tunnel backfill or the groundwater. Oxygen corrosion will occur initially and possibly intermittently during deglaciations. Microbially induced corrosion may have to be considered if the buffer deviates from its expected long-term behaviour.

Natural analogues/observations in nature

Corrosion of copper under oxidizing conditions can be studied on archaeological artefacts. Such studies have been conducted both to assess the risk of pitting /Bresle et al. 1983, Hallberg et al. 1984/. This was discussed above. A natural analogue studies assess general corrosion over longer periods of time is reported in /Hallberg et al. 1988/. /Hallberg et al. 1988/ examined a bronze cannon from the Swedish warship Kronan, which had lain buried with its muzzle downward in the mud on the bottom of the Baltic Sea since 1676 at a depth of 26 m. Microprobe analysis of a bronze core taken from the mussle showed concentrations of 96.3% Cu and 3.3% Sn and combined Zn and Fe < 0.5%. The dominant corrosion product was cuprite, Cu_2O. The estimated corrosion rate was $1.5 \cdot 10^{-5}$ mm/y with no evident pitting.

/Milodowski et al. 2003/ present an analysis of the corrosion of native copper plates that have survived in a water-saturated clay environment for more than 176 million years. Although the native copper is affected by corrosion, the study shows that a significant proportion (30–80% of the original thickness) of the copper sheets is preserved in the saturated compacted clay environment of the Littleham Mudstone. Apart from the recent weathering effects due to exposure at outcrop, petrographical studies demonstrate that most of the observed corrosion and alteration of the native copper is geologically old (i.e. predating the main sediment compaction) and also occurred before the end of the Lower Jurassic. This demonstrates that the native copper can remain stable in a saturated and compacted clay environment for geological timescales well in excess of the timescales considered for performance assessment of a deep geologic repository for spent nuclear fuel.

Handling in the safety assessment SR-Can

The process is modelled for the main scenario in SR-Can. The need for additional modelling in other scenarios will be evaluated. The model, that essentially consists of steady state mass balance and transport rate expressions, is presented in detail in /Hedin 2004/. The model is based on expressions that have been developed and applied to canister corrosion over several decades. The following contributions are considered in the modelling:

- Uneven general corrosion by initially entrapped oxygen, taking into account the amount of oxygen consumed by other processes in the buffer and backfill. This contribution is handled as a simple mass balance calculation. Limitations by transport rates of oxygen or reaction rates are disregarded.
  Corrosion by sulphide initially present in the buffer and supplied over time by transport in the groundwater. These contributions are modelled as transport-limited and any kinetic limitations due to interfacial reactions are disregarded.
- The likelihood and the quantitative effect of oxygen penetration during deglaciation will be assessed in dedicated model studies (see further the Process Report for the geosphere /SKB 2006h/). The corrosion model will be used to quantify corrosion by penetrating oxygen in the groundwater, using similar transport rate expressions to those employed for sulphide in the groundwater.
- Microbe mediated corrosion for a malfunctioning buffer will be treated by pessimistic assumptions.
- Chloride corrosion is neglected based on the discussion presented above.
- Nitric acid corrosion is neglected based on the discussion presented above.
- SCC of the copper canister is neglected (see further Section 3.5.5 below)

The canister's nominal copper coverage is 50 mm. The actual copper coverage, particularly in the weld regions will have a distribution of coverage less than that depending on the possible presence of flaws in the weld. Based on test results from the welding process it is assumed that 99% of all canisters have 40 mm copper coverage and the remaining 1% have 35 mm.

Boundary conditions: The boundary conditions, i.e. the rate of supply of corrosive agents from the near field to the canister – buffer boundary, are explicitly included in the model.

Handling of influences/couplings: Any temperature dependence of reaction rates are pessimistically neglected since all reactions are assumed to be instantaneous. The influence of the material properties is included through mass balance expressions and stochiometric formulae. The reduction in copper canister thickness is explicitly calculated.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There are no mechanistic uncertainties of any importance for the safety assessment. The extent of corrosion due to the residual oxygen in the repository is determined using a mass balance approach and the corrosion rate under reducing condition is determined by calculating the mass transport flux of corrosive agents to the canister surface. The mechanistic understanding of mass transport is sufficient for the needs of the safety assessment.

Model simplification uncertainties

The model simplifications are pessimistic; e.g. interfacial reaction kinetics are ignored and steady-state rather than transient transport rate expressions are employed.

Input data and data uncertainties

Input data and data uncertainties for the modelling of canister corrosion are discussed in the SR-Can Data Report /SKB 2006b/. Important input data include the amount of initially entrapped oxygen in the repository, diffusivities in the buffer, the solubility of sulphide in the buffer material, the initial amount of sulphide present in the buffer, the sulphide concentration in the groundwater over time, and the possible amounts of oxygen that could penetrate to repository depth during a deglaciation.

3.5.5 Stress corrosion cracking of the copper canister

Overview/general description

/King et al. 2001/ discuss in detail the current knowledge of SCC in copper and copper alloys and evaluate the risk for SCC of the copper canisters in the repository environment. This discussion has been further elaborated by /King and Kolar 2004, 2005/. In order for stress corrosion cracking to occur, a combination of several parameters will be required. The decisive ones are:

- The corrosion potential and the pH at the copper surface must be such that a duplex film of Cu_2O/CuO is thermodynamically stable.
- NH_3, NO_2^- or CH_3COO^- must be present at the copper surface durin the time the Cu_2O/CuO film is stable.
- The concentrations of NH_3, NO_2^- or CH_3COO^- must be sufficiently high in order to be able to induce stress corrosion cracking.
- The concentrations of Cl^- must be low in order not to inhibit stress corrosion cracking.
- Tensile stresses in the canister surface must be sufficiently high so that crack initiation and crack growth will happen.

Only early in the evolution of the repository will the necessary oxidants be available in sufficient concentrations to stabilise the Cu_2O/CuO film. During the water saturation phase, nitrogen compounds may be present os raiolysis products or residues from explosives. The concentrations that are foreseen are very low and will not constitute a risk for stress corrosion cracking, particularly since the copper canister before water saturation will (i.e. before the canister is under external overpressure) will be practically free of tensile stresses in the surface. After water saturation, tensile stresses can have occurred locally on the copper surface. The tensile stresses, however, are not present throughout the whole thickness of the copper canister /Cakmak 1994/. The residual oxygen will be consumed very rapidly in the repository, and the redox potential will be too low to stabilise the Cu_2O/CuO film. No tests in groundwater or simulated bentonite pore water have shown any signs of stress corrosion cracking in the copper material chosen for the canister /King et al. 2001/.

The following table shows how the process influences, and is influnced by, all canister variables.

Boundary conditions

Concentrations of nitrites, ammonium or acetate at the canister – buffer interface constitute the boundary conditions for this process.

Model studies/experimental studies

See Overview/general description.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes Lower temperatures increase the sensitivity to SCC.	Neglected The process is neglected as a failure mode for the canister.	No	
Canister geometry	No		No	
Material composition	Yes Different materials have different sensitivities to SCC.	Actual copper material is considered. The process is neglected as a failure mode for the canister.	No	
Mechanical stress	Yes Tensile stresses at the canister surface are a prerequisite for SCC.	Neglected The process is neglected as a failure mode for the canister.	Yes An SCC crack will relieve the tensile stresses.	

Time perspective

If SCC occurs, it will occur during an early phase in the repository when oxidizing conditions still prevail (< 300 years).

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

Based on the discussion above, the process is neglected as a failure mode for the canister.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There is no evidence that SCC could occur in the repository environment, but the possibility cannot be entirely ruled out based on available knowledge.

Tensile stresses in the copper canister are a necessary prerequisite, and since the canister is under external pressure it is very unlikely that SCC could lead to canister penetration. This would require tensile stresses through the whole canister wall, and such a situation is not deemed to be possible under the conditions in the repository.

Model simplification uncertainties

SCC is not modelled.

Input data and data uncertainties

Not relevant.

3.5.6 Earth currents – Stray current corrosion

Overview/general description

Sources of earth currents

There are several sources for electrical currents in the earth's crust, both natural and man made. One natural source is the stream of charged particles from the sun; the solar wind. Interaction between the solar wind and the earth's magnetic field causes currents to flow in the earth's crust. Another type of natural source is of an electrochemical nature. Oxidation of sulphide ores coupled to reduction of oxygen can give rise to a "battery-effect". Man-made sources include all types of electrical power installations. High Voltage Direct Current transmissions (HVDC) are locally very strong sources. Monopolar HVDC uses the ground to close the electrical circuit. Thousands of amperes are forced through the earth's crust over hundreds of kilometres. Although the electrodes are usually located in the sea and major parts of the current stay in the seawater, neighbouring land is usually affected.

Charge carriers

In water, the current is always carried by ions, e.g. Na^+ and Cl^-. Also in the ground, in clay, soil and rock, ions are the dominant charge carriers. Only certain minerals have significant electronic conductance. Although granite is a poor ion conductor, ionic current transport will dominate because it is an even poorer electronic conductor.

Alternating and direct currents

Depending on the source, earth currents are alternating or direct. Most man-made sources are alternating. In Sweden the usual frequency for power transmission is 50Hz. HVDC transmissions are, as the name implies, direct current. Although the source may be alternating, it is possible that a direct current component arises. A mineral with rectifying properties could, like a diode in a network of resistors, cause a DC component to arise in the vicinity of the rectifying mineral.

Corrosion effects on the canister caused by earth currents

Earth currents are known to cause corrosion. Primarily constructions stretching long distances, such as pipelines, are affected. Experience says that alternating currents do not generally cause corrosion. Corrosion effects have been observed only at very high AC densities.

In this context, a small metallic object such as a five meter high copper canister will be relatively insensitive to direct currents because the voltage drop across it will be low. Moreover, the repository is to be located in granitic rock with a low conductivity and the canister is to be embedded in highly conductive bentonite.

Metallic copper has negligible resistance and current transport must be electronic, while current in the bentonite will be ionic. Thus, conduction aross the copper-buffer interface can only be achieved if kinetically-facile reactions are available for interfacial charge transfer. The rate of such interfacial reactions is inversely proportional to the polarization resistance across the copper/buffer interface. As discussed in Section 3.5.4 the polarization resistances for copper corrosion are extremely large under anoxic conditions and at low sulphide concentrations. Thus, for a number of reasons the effects of earth currents on canister corrosion will be negligible. The surrounding granite ensures that the currents passing through the repository are low. The high conductivity of the bentonite and the large polarization resistances of the canister-buffer interfaces will ensure that the great majority of any earth currents through the repository will be transported through the buffer and only negligible amounts will be transported across the interfaces to cause canister corrosion. In short, the granite 'isolates' the repository and the bentonite is expected to conduct any repository current past the canister, leaving it unaffected.

The following table shows how the process influences, and is influenced by, all canister variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	No		No	
Canister geometry	Yes The dimensions of the canister determine its conductance.	The process is neglected	Yes To the extent that the process occurs, the effect would be a reduction of canister thickness.	The process is neglected
Material composition	Yes Different materials have different electrical conductivities.	The process is neglected	No	
Mechanical stress	No		No	

The possible variations and changes in canister geometry will have negligible consequences for this process.

Boundary conditions

The earth currents are generated in the far field and transferred to the canister at the canister – buffer boundary.

Model studies/experimental studies

Nothing is available at the time of SR-Can.

Time perspective

Earth currents generated by natural causes may be present throughout the one million year time period of the safety assessment. Man made earth currents are expected to disappear during glaciations.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process is neglected, based on the discussion presented above.

Handling of uncertainties in SR-Can

Uncertainties in mechanistic understanding

There are no uncertainties in the mechanistic understanding of any importance for the safety analysis.

Model simplification uncertainties for the above handling in SR-Can

Not applicable since the process is not modeled.

Input data and data uncertainties for the above handling in SR-Can

Not applicable since the process is not modeled.

3.5.7 Deposition of salts on canister surface

Overview/general description

In the KBS-3 safety assessment, the maximum surface temperature was calculated to be 80°C; i.e. well below the 100°C required to threaten the chemical stability of the bentonite could be assured /KBS-3 1983, Pusch 1983/. In the present design, the heat generation in the fuel is 1,700 W, and this will result in an increased canister surface temperature. The actual temperature will among other things depend on the heat transfer to the surrounding bentonite. At elevated temperatures, the corrosion reactions may proceed at an increased rate, but this will be of negligible importance since the corrosion is modelled under mass transport and mass balance control. A more important consequence of the higher canister surface temperature is the possible redistribution and enrichment of salts in the bentonite and their deposition onto the canister surface. The extent to which this may occur will depend on the salt content in the groundwater as well as in the bentonite. The salts that may be of concern are chlorides and sulphates from the groundwater and sulphates and carbonates from the impurities in the bentonite. Of these, the chlorides are most important since the presence of high chloride concentrations may have an effect on the corrosion properties of copper.

In the LOT experiment /Karnland et al. 2000/, copper heaters are buried in compacted bentonite. Two of these packages have been retrieved and analysed. These two heaters had surface temperatures of 90°C and 130°C, respectively. Examination of their surfaces showed that in both cases the copper surfaces were covered with a thin layer of calcium sulphate/calcium carbonate. No chloride enrichments were apparent even though the groundwater had a chloride content of over 8,000 mg/L. To date it has not been established whether the precipitation of the sulphates and the carbonates was caused by evaporation of water or by the lower solubility of the calcium salts at elevated temperatures. If the latter is the case, they are likely to redissolve as the temperature decreases. Similar salt deposits were observed in a heater experiment performed in Stripa /Pusch et al. 1992/.

It is at this time not established whether or not the precipitation of the sulphates and the carbonates were caused by evaporation of water or by the lower solubility of the calcium salts at elevated temperatures. In the latter case, they are likely to redissolve as the temperature decreases again. Alternatively, the deposits can be redissolved when contacted with water undersaturated with respect to calcium carbonate and calcium sulphate. Even if this surface cover remains over longer periods of time, it is unlikely that any credit can be taken for it having protective properties. The deposits are not electrically conductive, so they are not expected to increase the risk of pitting corrosion, see e.g. /Adeloju and Duan 1994/. Also, an increase in the chloride concentration would lower the susceptibility of copper for pitting corrosion, since it would favour general corrosion /King et al. 2001/. A high chloride concentration will, however, not lead to increased general corrosion, since the near field pH is always expected to be slightly alkaline and, consequently, the extent of the corrosion will be determined by the amount of available oxygen.

Thus, exposures to temperatures over 100°C is not expected to have an effect on the corrosion behaviour of the copper canister. The maximum allowed surface temperature of the canister is, therefore, set more by the requirements on chemical stability of the bentonite buffer than by any possible influence on canister corrosion. Cu_2O/CuO film.

The following table shows how the process influences, and is influenced by, all canister variables.

Variable	Variable influence on process		Process influence on variable	
	Influence present?	Handling of influence	Influence present?	Handling of influence
Radiation intensity	No		No	
Temperature	Yes See discussion above.	Neglected The process has no implications for safety.	No	
Canister geometry	No		No	
Material composition	No		Yes Surface deposites may occur.	Neglected The process has no implications for safety.
Mechanical stress	No		No Negligible stresses will be induced in the canister.	

Boundary conditions

The heat is generated in the canister and transferred to the canister buffer boundary.

Model studies/experimental studies

See above.

Time perspective

Elevated temperatures will remain for several hundred years. The effects discussed above will, however, only occur during the bentonite saturation phase.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The process has no implications for safety and is therefore ignored.

Handling of uncertainties in SR-Can

Not applicable.

Uncertainties in mechanistic understanding

There are uncertainties concerning the mechanisms for possible salt deposition. There are no major uncertainties in understanding the consequences of such a deposit on the lifetime of the canister.

Model simplification uncertainties for the above handling in SR-Can

Not applicable.

Input data and data uncertainties for the above handling in SR-Can

Not applicable.

3.6 Radionuclide transport

See Section 2.6 “Radionuclide transport “.

4 References

Aaltonen P, Varis P, 1993. Long term corrosion tests of OFHC-coppers in simulated repository conditions – final report. YJT-93-05. Nuclear Waste Commission of Finnish Power Companies Report.

Adeloju S B, Duan Y Y, 1994. Influence of carbonate ions on stbility of copper oxides and copper pitting corrosion, Br. Corr. J. 29, p.315.

Agrenius L, 2002. Criticality safety calculations of storage canisters. SKB TR-02-17, Svensk Kärnbränslehantering AB.

Albinsson Y, Ödegaard-Jensen A, Oversby V M, Werme L, 2003. Leaching of spent fuel under anaerobic and reducing conditions. In: Scientific Basis for Nuclear waste management XXVI, Mat. Res. Soc. Symp. Series, vol. 757, (Finch, R. J. and Bullen, D.B. eds.), pp 407–413,

Allen A O, 1961. The radiation chemistry of water and aqueous solutions. Princeton (USA) D.Van Nostrand Co. Inc.

Andersson C-G, Andersson M, Björkegren L-E, Dillström P, Erixon B, Minnebo P, Nilsson F, Nilsson K-F, 2005. Probabilistic Analysis and Materials Characterisation of Canister Insert for Spent Nuclear Fuel – Summary Report. SKB TR-05-17. Svensk Kärnbränslehantering AB.

Beavers J A, Thompson N G, Parkins R N, 1985. Stress corrosion cracking of low strength carbon steels in candidate high level waste repository environments: environmental effects. Nuclear and Chemical Waste Management 5, pp 279–296.

BEFAST III, 1997. Further analysis of extended storage of spent fuel. Final report of a Co-ordinated Research Programme on the Behaviour of Spent Fuel Assemblies during Long-term Storage (BEFAST III) 1991–1996. IAEA TECDOC-944.

Berner U, Curti E, 2002. Radium solubilities for SF/HLW wastes using solid solution and co-precipitation models. Internal Report TM-44-02-04, PSI, Villigen, Switzerland.

Blackwood D J, Naish C C, 1994. The effect of galvanic coupling between the copper outer canister and the carbon steel inner canister on the corrosion resistance of the Advanced Cold process canister. SKB Inkapsling PR 95-04, Svensk Kärnbränslehantering AB.

Blackwood D J, Henshaw J, Platts N, Hilditch J P, 1995. Stress corrosion cracking of the advanced cold process canister: Carbon steel in nitric acid vapour. SKB Inkapsling PR 96-05, Svensk Kärnbränslehantering AB.

Bond A E, Hoch A R, Jones G D, Tomczyk A J, Wiggin R W, Worraker W J, 1997. Assessment of a spent fuel disposal canister. Assessment studies for a copper canister with cast steel inner component. SKB TR 97-19, Svensk Kärnbränslehantering AB.

Brantberger M, Zetterqvist A, Arnbjerg-Nielsen T, Olsson T, Outters N, Syrjänen P, 2006. Final repository for spent nuclear fuel. Underground design Forsmark, Layout D1. SKB R-06-34, Svensk Kärnbränslehantering AB.

Bresle Å, Saers J, Arrhenius B, 1983. Studies on pitting corrosion on Archaelogical bronzes. SKB TR 83-05, Svensk Kärnbränslehantering AB.

Bruno J, Cera E, de Pablo J, Duro L, Jordana S, Savage D, 1997. Determination of radionuclide solubility limits to be used in SR 97. Uncertainties associated to calculated solubilities. SKB TR 97-33, Svensk Kärnbränslehantering AB.

Börgesson L, 1986. Model shear tests of canisters with smectite clay envelopes in deposition holes. SKB TR 86-26, Svensk Kärnbränslehantering AB.

Börgesson L, Hernelind J, 1998. Uneven swelling pressure on the canister. FEM-calculation of the effect of uneven water supply in the rock. SKB Projekt Inkapsling PPM 98-3420-33.

Börgesson L, Johannesson L-E, Hernelind J, 2004. Earthquake induced rock shear through deposition hole. Effect on the canister and the buffer. SKB TR-04-02. Svensk Kärnbränslehantering AB.

Börgesson L, Hernelind J, 2006. Earth quake induced rock shear through a deposition hole. Influence of shear plane inclination and location as well as buffer properties on the damage caused to the canister. SKB TR-06-XX (in press), Svensk Kärnbränslehantering AB.

Cakmak E, 1994. Beräkningar av maximal töjning i kopparbehållare för slutförvaring av utbränt kärnbränsle. SKB Inkapsling PPM 95-3420-01, Svensk Kärnbränslehantering AB. (In Swedish).

Carbol P, Cobos-Sabate J, Glatz J-P, Grambow B, Kienzler B, Loida A, Martinez Esparza Valiente A, Metz V, Quiñones J, Ronchi C, Rondinella V, Spahiu K, Wegen D H, Wiss T, 2005. The effect of dissolved hydrogen on the dissolution of ^{233}U doped UO_2(s), high burnup spent fuel and MOX fuel, *SKB Technical Report, In preparation.*

Christensen H, Bjergbakke E, 1982. Radiolysis of groundwater from HLW stored in copper canisters. SKBF/KBS TR 82-02. Svensk Kärnbränslehantering AB.

Christensen H, Bjergbakke E, 1984. Radiolysis of concrete. SKBF/KBS TR 84-02. Svensk Kärnbränslehantering AB.

Christensen H, Pettersson S-O, 1997. Beräkning av radiolys utanför intakt kapsel, Studsvik Material, Report STUDSVIK/M-97/67.

Cliffe K A, Kelly M, 2005. COMP23 Version 1.2.2 User's Manual. SKB R-04-64, Svensk Kärnbränslehantering AB.

Cui D, Spahiu K, **2002.** The reduction of U(VI) on corroded iron under anoxic groundwater conditions, Radiochim. Acta, 90, pp 1–6.

Cui D, Low J, Sjöstedt C J, Spahiu K, 2004. On Mo-Ru-Tc-Pd-Rh alloy particles extracted from spent fuel and their leaching behavior under repository conditions. Radiochim. Acta, 94 (2004) 551–555.

Desgranges L, Ripert M, Piron J P, Kodja H, Gallier J P, 2003. Behaviour of fission gases in an irradiated nuclear fuel under α external irradiation, J. Nucl. Mater. 321, pp 324–330.

Dillström P, 2005. Probabalistic analysis of canister inserts for spent nuclear fuel**.** SKB TR-05-19. Svensk Kärnbränslehantering AB.

Doerner H A, Hoskins W M, 1925. Coprecipitation of barium and radium sulphates, J. Am. Chem. Soc. 47, 662–675.

Duro L, Grivé M, Cera E, Domenech C, Bruno J, 2004. Update of a thermodynamic database for radionuclides to assist solubility limit calculations, Enviros report.

Duro L, Grivé M, Cera E, Gaona X, Domènech C, Bruno J, 2005. Determination and assessment of the concentration limits to be used in SR-Can. SKB TR-05-?? Svensk Kärnbränslehantering AB.

EN 1563:1997. Founding – Spheroidal graphite cast iron.

EN 1976:1988. Copper and copper alloys – Cast unwrought copper products. (UNS10100: Copper Development Association Inc. Application Data Sheet. Standard designations for copper and copper alloys (www.copper.org)).

Eriksen T, 1996. Radiolysis of water within a ruptured fuel element. SKB PR U-96-29, Svensk Kärnbränslehantering AB.

Evins L Z, Jensen K A, Ewing R C, 2005. Uraninite recrystallization and Pb loss in the Oklo and Bangombé natural fission reactors, Gabon. Geochim. Cosmochim. Acta, 69, pp 1589–1606.

Forsyth R S, Mattsson O, Schrire D, 1988. Fission product concentration profiles (Sr, Xe, Cs and Nd) at the individual grain level in power-ramped LWR fuel. SKB TR 88-24, Svensk Kärnbränslehantering AB.

Forsyth R S, Jonsson T, Mattsson O, 1990. Examination of reaction products on the surface of UO_2 fuel exposed to reactor coolant water during power operation. SKB TR 90-07, Svensk Kärnbränslehantering AB.

Forsyth R S, Werme L O, 1992. Spent fuel corrosion and dissolution. J. Nucl. Mater. pp 190, 3–19.

Forsyth R S, Eklund U-B, Werme L O, 1994. A study of fission product migration and selective leaching by meand of a power-bump test. Mat. Res. Soc. Symp. Proc. Vol. 333, 385–390.

Forsyth R, 1995. Spent nuclear fuel. A review of properties of possible relevance to corrosion processes. SKB TR 95-23, Svensk Kärnbränslehantering AB.

Forsyth R, 1997. The SKB Spent Fuel Corrosion Programme. An evaluation of results from the experimental programme performed in the Studsvik Hot Cell laboratory. SKB TR 97-25, Svensk Kärnbränslehantering AB.

Gdowski G E, Bullen D B, 1988. Survey of degradation modes of candidate materials for high-level radioactive-waste disposal containers. Oxidation and corrosion. Report UCID-21362 Vol. 2. Lawrence Livermore National Laboratory, University of California, Livermore, California, USA.

Grambow B, Loida A, Dressler P, Geckeis H, Gago J, Casas I, de Pablo J, Gimenez J, Torrero M E, 1996. Long-term safety of radioactive waste disposal: Chemical reaction of fabricated and high burnup spent fuel with saline brines, Forschungszentrum Karlsruhe, Wissenschaftliche Berichte FZKA 5702.

Grambow B, Loida A, Martinez-Esparza A, Diaz-Arocas P, de Pablo J, Paul J-L, Marx G, Glatz J-P, Lemmens K, Ollila K, Christensen H, 2000. Source term for performance assessment of spent fuel as a waste form, European Commission, Nuclear Science and Technology, EUR 19140 EN 2000.

Grandia F, Domènech C, Arcos D, Duro L, 2006. Assessment of the oxygen consumption in the backfill. Geochemical modelling in a saturated backfill. SKB TR-06-YY, Svensk Kärnbränslehantering AB.

Grauer R, 1991. The reducibility of sulphuric acid and sulphate in aqueous solution (translated from German). SKB TR 91-39, Svensk Kärnbränslehantering AB.

Gray W J, Strachan D M, Wilson C N, 1992. Gap and grain boundary inventories of Cs, Tc and Sr in spent LWR fuel, Mat. Res. Soc. Symp. Proc. Vol. 257, 353–360.

Gray W J, 1999. Inventories of iodine-129 and cesium-137 in the gaps and grain boundaries of LWR fuels, Mat. Res. Soc. Symp. Proc. Vol. 556, 487–494.

Grenthe I, Fuger J, Konings R J M, Lemire R, Muller A B, Nguen - Trung C, Wanner H, 1992. Chemical Thermodynamics of Uranium. Elsevier Science Publishers, Amsterdam.

Grigoriev V, 1996. Hydrogen embrittlement of zirconium alloys. Studsvik Material, Report STUDSVIK/M-96/73.

Guilbert S, Sauvage T, Erramli H, Barthe M-F, Desgardin P Blondiaux G, Corbel C, Piron J P, 2003. Helium behavior in UO_2 polycrystalline disks, J. Nucl. Mater. 321, pp 121–128.

Guillaumont R, Fanghänel T, Fuger J, Grenthe I, Neck V, Palmer D A, Rand M H, 2003. Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium**.** Elsevier Science Publishers, Amsterdam.

Guinan M W, 2001. Radiation effects in spent nuclear fuel canisters. SKB TR-01-32, Svensk Kärnbränslehantering AB.

Hallberg R, Engvall A-G, Wadsten T, 1984. Corrosion of copper lightning conductor plates. Br. Corros. J. Vol. 19:2, pp 85–88.

Hallberg R O, Östlund P, Wadsten T, 1988. Inferences from a corrosion study of a bronze cannon, applied to high level nuclear waste disposal. Applied Geochemistry, Vol. 3, pp 273–280.

Hedin A, 2004. Integrated near-field evolution model for a KBS-3 repository. SKB R-04-36, Svensk Kärnbränslehantering AB.

Henshaw J, Hoch A, Sharland S M, 1990. Further assessment studies of the Advanced Cold Process Canister. AEA Decommissioning & Radwaste, AEA D&R 0060.

Henshaw J, 1994. Modelling of nitric acid production in the Advanced Cold Process Canister due to irradiation of moist air. SKB TR 94-15, Svensk Kärnbränslehantering AB.

Hernelind J, 2006. Earthquake induced rock shear through a deposition hole when creep is considered – first model. Effect on the canister and the buffer. SKB R-06-87, Svensk Kärnbränslehantering AB.

Hocking W H, Duclos A M, Johnson L H, 1994. Study of fission-product segregation in used CANDU ffuel by X-ray photoelectron spectroscopy (XPS) II. J. Nucl. Mater. 209, 1–26.

Håkansson R, 1995. Dosratsberäkningar för kapsel med använt kärnbränsle. Studsvik Nuclear AR N(R)-95/026. (In Swedish).

Håkansson R, 1999. Beräkning av nuklidinnehåll, resteffekt, aktivitet samt doshastighet för utbränt kärnbränsle. SKB R 99-74, Svensk Kärnbränslehantering AB. (In Swedish).

Janson T, Magnusson J, Bergvall M, Olsson R, Cuisat F, Skurtveit E, Grimstad E, 2006. Final repository for spent ffuel. Underground design Laxemar, Layout D1. SKB R-06-36, Svensk Kärnbränslehantering AB.

Johnson L H, Leneveu D M, King F, Shoesmith D W, Kolar M, Oscarson D W, Sunder S, Onofrei C, Crosthwaite J L, 1996. The disposal of Canada's nuclear fuel waste: A study of post-closure safety of in-room emplacement of used CANDU fuel in copper containers in permeable plutonic rock. Volume 2: Vault model. Atomic Energy of Canada Limited Report, AECL-11494-2, COG-95-552-2.

Johnson L H, Tait J C, 1997. Release of segregated nuclides from spent fuel. SKB TR 97-18, Svensk Kärnbränslehantering AB.

Johnson L H, McGinnes D F, 2002. Partitioning of radionuclides in Swiss power reactor fuels, Nagra Technical report 02-07.

Johnson L H, Poinssot C, Ferry C, Lovera P, 2004. Esimates of the instant release fraction of UO_2 and MOX fuel at t = 0. NAGRA Technical Report 04-08.

Kamikura K, 1992. FP gas release behaviour of high burnup MOX fuels for thermal reactors, Proc. of Technical Committee Meeting on Fission Gas Release and Fuel Rod Chemistry Related to Extended Burnup, Pembroke, Ontario, Canada, 28 April – 1 May 1992, IAEA-TECDOC-697, p 82.

Karnland O, Sandén T, Johannesson L-E, Eriksen T E, Jansson M, Wold S, Pedersen K, Motamedi M, Rosborg B, 2000. Long term test of buffer material. Final report on the pilot parcels. SKB TR-00-22, Svensk Kärnbränslehantering AB.

Kelm M, Bohnert E, 2004. A kinetic model for the radiolysis of chloride brine, its sensitivity against model parameters and a comparison with experiments, FZKA 6977 Report, Forschungszentrum Karlsruhe.

King F, LeNeveu D, 1992. Prediction of the lifetimes of copper nuclear waste containers. In Proceedings of Conference on Nuclear Waste Packaging, FOCUS '91, (American Nuclear Society, La Grange Park, IL), 253–261.

King F, Litke C D, Ryan S R, 1992. A mechanistic study of the uniform corrosion of copper in compacted Na-montmorillonite/sand mixtures, Corros. Sci. 33, pp 1979–1995.

King F, Kolár M, 1997. The effect of geosphere conditions on the lifetimes of copper containers, AECL-11717, COG-96-565. Atomic Energy of Canada Limited.

King F, Kolár M, 2000. The copper container corrosion model used in AECL's second case study. 06819-REP-01200-10041-R00. Ontario Power Generation, Nuclear Waste Management Division.

King F, Ahonen L, Taxén C, Vuorinen U, Werme L, 2001. Copper corrosion under expected conditions in a deep geologic repository, SKB Technical Report TR-01-23, Svensk Kärnbränslehantering AB (also available as Posiva Report 2002-01 (2002)).

King F, Kolar M, 2004. Theory manual for the copper corrosion model for stress corrosion cracking of used fuel disposal containers CMM-SCC.0, Ontario Power Generation Report No: 06819-REP-01300-10095-R00.

King F, Kolar M, 2005. Preliminary assessment of the stress corrosion cracking of used fuel disposal containers using the CMM-SCC.0 model, Ontario Power Generation Report No: 06819-REP-01300-10103-R00.

King F, Shoesmith D W, 2004. Electrochemical studies of the effect of H_2 on UO_2 dissolution. SKB TR-04-20, Svensk Kärnbränslehantering AB.

KBS-3: Final Storage of Spent Nuclear Fuel – KBS-3, Part III Barriers (p. 9:6), 1983. SKBF/KBS, Svensk Kärnbränsleförsörjning AB.

Lemire R J, Fuger J, Nitsche H, Potter P, Rand M H, Rydberg J, Spahiu K, Sullivan J C, Ullman W J, Vitorge P, Wanner H, 2001. Chemical Thermodynamics of Neptunium and Plutonium. Elsevier Science Publishers, Amsterdam.

Litke C D, Ryan S R, King F, 1992. A mechanistic study of the uniform corrosion of copper in compacted clay-sand soil, AECL-10397, COG-91-304. Atomic Energy of Canada Limited.

Lovera P, Ferry C, Poinssot C, Johnson L, 2003. Synthesis report on the relevant diffusion coefficients of fission products and helium in spent nuclear fuels. Report CEA-R-6039, Commisariat à l'Énergie Atomique.

Loida A, Grambow B, Geckeis H, 1996. Anoxic corrosion of various high burnup spent fuel samples, J. Nucl. Mater., 238, pp. 11–22.

Lundgren K, 2004. Final disposal of fuel – electron radiation outside copper canister. SKB TR-04-06, Svensk Kärnbränslehantering AB.

Marion G M, Kargel J S, Catling D C, Jakubowski S D, 2005. Effect of pressure on aqueous chemical equilibria at subzero temperatures with applications to Europa, Geochim. Cosmochim Acta, 69, 259–274.

Marsh G P, 1990. A preliminary assessment of the advanced cold process canister. AEA Technology, Report AEA-InTec-0011.

Masurat P, 2006. Potential for corrosion ín disposal systems for high level waste by *Meiothermus* and *Desulfovibrio.* Ph. D. Thesis, Göteborg UniversityISBN 91-628-6773-3.

Masurat P, Pedersen K, 2004. Microbial sulphide production in compacted bentonite at the commencement of long-term disposal of high-level radioactive waste, Mat. Res. Soc. Symp. Proc. Vol. 807, pp 805–810.

Matthiesen H, Hilbert L R, Gregory D, 2003. Siderite as a corrosion product on archaeological iron from a waterlogged environment. Studies in Conservation vol. 48, pp 183–194.

Matthiesen H, Hilbert L R, Gregory D, Sørensen B, 2004. Corrosion of archaeological iron artefacts compared to modern iron at the waterlogged site Nydam, Denmark., Prediction of Long Term Corrosion Behaviour in Nuclear Waste System. Proceedings of the 2nd International Workshop organized by the Working Party on nuclear corrosion (WP4) of the European Federation of Corrision (EFC), Nice, September 2004, Eurocorr 2004, Ed. by Andra, pp. 114–127.

Mattsson H, Olefjord I, 1984. General corrosion of Ti in hot water and water saturated bentonite clay. SKB/KBS TR 84-19.

Mattsson H, Li C, Olefjord I, 1990. Analysis of oxide formed on titanium during exposure in bentonite clay. II. The structure of the oxide. Werkstoffe und Korrosion 41, 578–584.

Mattsson H, Olefjord I, 1990. Analysis of oxide formed on titanium during exposure in bentonite clay. I. The oxide growth. Werkstoffe und Korrosion 41, 383–390.

Milodowski A E, Styles M T, Werme L, Oversby V M, 2003. The corrosion of more than 176 million year**,** old native copper plates from a deposit in mudstone in south devon; united kingdom, NACE Corrosion 2003, Paper No. 03681.

Motamedi M, Karnland O, Pedersen K, 1996. Survival of sulfate reducing bacteria at different water activities in compacted bentonite. FEMS Microbiology Letters 141, pp 83–87.

Neff D, Dillmann P, Beranger G, 2003. An analytical study of corrosion products formed on buried ferrous archaeological artefacts. In: Prediction of long term corrosion behaviour in nuclear waste systems, eds. D. Féron and D.D. Macdonald. EFC publication 36, pp 295–315.

Neff D, Dillmann P, Bellot-Gurlet L, Beranger G, 2005. Corrosion of iron archaeological artefacts in soil: characterisation of the corrosion system. Corrosion Science, vol. 47, pp 515–535.

Neff D, Dillmann P, Descostes M, Beranger G, 2006. Corrosion of iron archaeological artefacts in soil: Estmation of the average corrosion rates involving analytical techniques and thermodynamic calculations. Corrosion Science vol. 48, pp 2947–2970.

Nilsson K-F, Burström M, Lofaj F, Andersson C-G, 2005. Pressure tests of two KBS-3 canister mock-ups. SKB TR-05-18, Svensk Kärnbränslehantering AB.

Northwood D, Kosasih U, 1983. Hydrides and delayed hydrogen cracking in zirconium and its alloys. Int. Metals Reviews 28, 92–121.

NUREG/CR 0200 SCALE documentation.

Olander D, 2004. Thermal spike theory of athermal diffusion of fission products due to alpha decay of actinides in spent fuel (UO_2). SKB TR-04-17, Svensk Kärnbränslehantering AB.

Ollila K, Albinsson Y, Oversby V, Cowper M, 2003. Dissolution rates of unirradiated UO_2, UO_2 doped with ^{233}U, and spent fuel under normal atmospheric conditions and under reducing conditions using an isotope dilution method. SKB TR-03-13, Svensk Kärnbränslehantering AB.

Ollila K. Oversby V M, 2005. Dissolution of unirradiated UO_2 and UO_2 doped with ^{233}U under reducing conditions. SKB TR-05-07, Svensk Kärnbränslehantering AB.

Oversby V M, 1996. Criticality in a high level waste repository. A review of some important factors and an assessment of the lessons that can be learned from the Oklo reactor. SKB TR 96-07, Svensk Kärnbränslehantering AB.

Oversby V M, 1998. Criticality in a repository for spent fuel: lessons from Oklo. Mat. Res. Soc. Symp. Proc. Vol. 506, p 781.

Pastina B, Isabey J, Hickel B, 1999. The influence of water chemistry on the radiolysis of primary coolant water in pressurized water reactors, J. Nucl. Mat., 264, pp 309–318.

Pastina B, LaVerne J A, 2001. Effect of molecular hydrogen on hydrogen peroxide in water radiolysis, J. Phys. Chem. A., 105, pp 9316–9322.

Pedersen K, Motamedi M, Karnland O, 1995. Survival of bacteria in nuclear waste buffer materials. The influence of nutrients, temperature and water activity. SKB TR 95-27, Svensk Kärnbränslehantering AB.

Pedersen K, Motamedi M, Karnland O, Sandén T, 2000. Mixing and sulphate-reducing activity of bacteria in swelling, compacted bentonite clay under high-level radioactive waste repository conditions, Journal of Applied Microbiology 89, pp 1038–1047.

Plan 2005. Kostnader för kärnkraftens radioaktiva restprodukter, 2005. Svensk Kärnbränslehantering AB.

Poinssot C, Toulhoat P, Piron J-P, Cappelare C, Desgranges L, Gras J-M, 2000. Operational and scientific questions related to the long term evolution of spent nuclear fuel in dry conditions. Current ststus of the French research, Proc. Embedded Topical Meeting on DOE Spent Nuclear Fuel and Fissile Material Management, San Diego, pp 3–10, American Nuclear Society, La Grange Park, IL.

Poinssot C, Jegou C, Toulhoat P, Piron J-P, Gras J-M, 2001. A new approach to the RN source term for spent nuclear fuel under geological disposal conditions, Mat. Res Soc. Symp. Proc. Vol. 663, pp 469–476.

Poinssot C, Lovera P, Faure M-H, 2002. Assessment of the evolution with time of the instant release fraction of spent nuclear fuel in geological disposal conditions, Mat. Res. Soc. Symp. Proc. Vol. 713, pp 615–623.

Pusch R, 1983. Stability of deep-sited smectite minerals in crystalline rock – chemical aspects. TR 83-16, Svensk Kärnbränsleförsörjning AB/Avdelning KBS.

Pusch R, Börgesson L, 1992. PASS – Project on alternative systems study. Performance assessment of bentonite clay barrier in three repository concepts: VDH, KBS-3 and VLH. SKB TR 92-40, Svensk Kärnbränslehantering AB.

Pusch R, Karnland O, Lajudie A, Lechelle J, Bouchet A, 1992. Hydrothermal field test with French candidate clay embedded steel heater in the Stripa mine. SKB TR 93-02, Svensk Kärnbränslehantering AB.

Rard J A, Rand M H, Anderegg G, Wanner H, 1999. Chemical Thermodynamics of Technetium. Elsevier Science Publishers, Amsterdam.

Rai D, Yui M, Moore D A, 2003. Solubility and Solubility Product at 22°C of UO_2(c) Precipitated From Aqueous U(IV) Solutions, J. Sol. Chem. Vol. 32, pp 1–17.

Reed D T, van Konynenburg R A, 1991a. Effect of ionizing radiation on the waste package environment. High Level Waste Management II, American Nuclear Society, La Grange Park, IL, 1396–1403.

Reed D T, van Konynenburg R A, 1991b. Progress in evaluating the corrosion of candidate HLW container metals in irradiated air-steam mixtures. Proceedings Nuclear Waste Packaging, Focus '91, American Nuclear Society, La Grange Park, IL, 185–192.

Romanoff M, 1989. Underground Corrosion. NACE International.

Roos A, 2003. The emissivity of copper from electron beam welds. Letter report to Lars Werme, Uppsala University October 2003.

Rothman A J, 1984. Potential corrosion and degradation mechanisms of Zircaloy cladding on spent fuel in a tuff repository. Lawrence Livermore National Laboratory report UCID-20172.

Roudil D, Deschanels X, Trocellier P, Jégou C, Peuget S, Bart J-M, 2004. Helium thermal diffusion in a uranium dioxide matrix, J. Nucl. Mater. 325, pp 148–158.

Scott T B, Allen G C, Heard P J, Randel M G, 2005. Reduction of U(VI) to U/IV) on the surface of magnetite, Geochim. Cosmochim. Acta, 69, pp 5639–5646.

Silva R J, Bidoglio G, Rand M H, Robouch P B, Wanner H, Puigdomenech I, 1995. Chemical Thermodynamics of Americium. Elsevier Science Publishers, Amsterdam.

SKB, 1999. SR 97 – Processes in the repository evolution. Background report to SR 97. SKB TR-99-07, Svensk Kärnbränslehantering AB.

SKB, 2003. Plan 2003. Costs for management of the radioactive waste products from, nuclear power production. SKB TR-03-11, Svensk Kärnbränslehantering AB.

SKB, 2004a. RD&D-Programme 2004. Programme for research, development and demonstration of methods for the management and disposal of nuclear waste, including social science research. SKB TR-04-21, Svensk Kärnbränslehantering AB.

SKB, 2004b. Interim Main report of the safety assessment SR-Can. SKB TR 04-11, Svensk Kärnbränslehantering AB.

SKB, 2005. Preliminary site description. Forsmark area - version 1.2. SKB R-05-18, Svensk Kärnbränslehantering AB.

SKB, 2006a. Long-term safety for KBS-3 repositories at Forsmark and Laxemar – a first evaluation. Main report of the SR-Can project. SKB TR-06-09, Svensk Kärnbränslehantering AB.

SKB, 2006b. Data report for the safety assessment SR-Can. SKB TR-06-25, Svensk Kärnbränslehantering AB.

SKB, 2006c. FEP report for the safety assessment SR-Can. SKB TR-06-20, Svensk Kärnbränslehantering AB.

SKB, 2006d. Initial State report for the safety assesment SR-Can. SKB TR-06-21, Svensk Kärnbränslehantering AB.

SKB, 2006e. Climate and climate related issues for the safety assessment SR-Can. SKB TR-06-23, Svensk Kärnbränslehantering AB.

SKB, 2006f. Handling of future human actions in the safety assessment SR-Can. SKB TR-06-24, Svensk Kärnbränslehantering AB.

SKB, 2006g. Buffer and backfill process report for the safety assessment SR-Can, SKB TR-06-18, Svensk Kärnbränslehantering AB.

SKB, 2006h. Geosphere process report for the safety assessment SR-Can. SKB TR-06-19, Svensk Kärnbränslehantering AB.

SKB, 2006i. Biosphere process report for the safety assessment SR-Can. SKB TR-06-XX, Svensk Kärnbränslehantering AB.

SKB, 2006j. Preliminary site description. Laxemar subarea - version 1.2. SKB R-06-10, Svensk Kärnbränslehantering AB.

Smart N R, Blackwood D J, Werme L, 2002a. Anaerobic corrosion of carbon steel and cast iron in artificial groundwaters: Part 1 – Electrochemical aspects. Corrosion 58, pp 547–559.

Smart N R, Blackwood D J, Werme L, 2002b. Anaerobic corrosion of carbon steel and cast iron in artificial groundwaters: Part 2 – Gas generation. Corrosion 58, pp 627–637.

Smart N R, Rance A P, Fennell, P, Werme L, 2003. Expansion due to anaerobic corrosion of steel and cast iron: experimental and natural analogue studies. In: Prediction of long term corrosion behaviour in nuclear waste systems, eds. D. Féron and D.D. Macdonald. EFC publication 36, pp 280–294.

Smart N R, Rance A P, Fennell, P A H, 2005. Galvanic corrosion of copper-cast iron couples. SKB TR 05-06. Svensk Kärnbränslehantering AB.

Smart N R, Fennell P A H, Rance A P, 2006. Expansion due to anaerobic corrosion of iron. SKB TR 06-41. Svensk Kärnbränslehantering AB.

Spinks J W T, Woods R J, 1990. An introduction to radiation chemistry. 3rd edition. New York, NY (USA). John Wiley and Sons Inc

Stroes-Gascoyne S, 1996. Measurements of instant-release source terms for ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{129}I and ^{14}C in used CANDU fuels, J. Nucl. Mater. 238, 265–277.

Swedish Corrosion Institute, 1978. Copper as canister material for unreprocessed nuclear waste − evaluation with respect to corrosion, KBS Teknisk Rapport 90, Kärnbränslesäkerhet.

Swedish Corrosion Institute, 1983. Corrosion resistance of a copper canister for spent nuclear fuel, SKBF/KBS TR 83-24, Svensk Kärnbränsleförsörjning AB.

Thomas L E, Charlot L A, Engelhard M H, 1988. Auger analysis of grain boundaries in LWR spent fuel. Pacific Northwest Laboratory, opublicerade resultat.

Thomas L E, Guenther R J, 1989. Characterization of low-gas-release LWR fuels by transmission electron microscopy. Mat. Res. Symp. Proc. Vol. 127, 293–300. Materials Research Society, Pittsburgh, USA.

Van Brutzel L, Rarivomanantsoa M, Ghaleb D, 2006. Displacement cascade initiated with the realistic energy of the recoil nucleus in UO_2 matrix by molecular dynamics simulation. J. Nucl. Mater. 354, pp 28–35.

van Konynenburg R A, 1994. Behavior of carbon-14 in waste packages for spent fuel in a tuff repository. Waste Management, v. 14, 363–383.

Werme L, Sellin P, Kjellbert N, 1992. Copper canisters for nuclear high level waste disposal. Corrosion aspects. SKB Technical Report TR-92-26, Svensk Kärnbränslehantering AB.

Werme L O, Forsyth R S, 1988. Spent UO_2 fuel corrosion in water; Release mechanisms. Mat. Res. Soc. Symp. Proc. Vol. 112, pp 443–452.

Werme L O, 1998. Design premises for canister for spent nuclear fuel. SKB TR-98-08, Svensk Kärnbränslehantering AB.

Werme L O, Spahiu K, 1998. Direct disposal of spent nuclear fuel: Comparison between experimental and modelled actinide solubilities in natural waters. J. Alloys and Compounds 271–273, 194–200.

Werme L O, Johnson L H, Oversby V M, King F, Spahiu K, Grambow B, Shoesmith D W, 2004. Spent fuel performance under repository conditions: A model for use in SR-Can, SKB TR 04-19, Svensk Kärnbränslehantering AB.

Wersin P, Bruno J, Spahiu K, 1993. Kinetic modelling of bentonite-canister interaction. Implications for Cu, Fe and Pb corrosion in a repository for spent nuclear fuel. SKB TR 93-16, Svensk Kärnbränslehantering AB.

Wersin P, Spahiu K, Bruno J, 1994a. Time evolution of dissolved oxygen and redox conditions in a HLW repository for spent nuclear fuel. SKB TR 94-02, Svensk Kärnbränslehantering AB.

Wersin P, Spahiu K, Bruno J, 1994b. Kinetic modelling of bentonite-canister interaction. Long-term predictions of copper canister corrosion under oxic and anoxic conditions. SKB TR 94-25, Svensk Kärnbränslehantering AB.

Wikramaratna R S, Goodfield M, Rodwell W R, Nash P J, Agg P J, 1993. A preliminary assessment of gas migration from the copper/steel canister. SKB TR 93-31, Svensk Kärnbränslehantering AB.

Wilson C N, 1990a. Results from NNWSI series bare fuel dissolution tests. Pacific Northwest Laboratory, Report PNL-7169, UC-802.

Wilson C N, 1990b. Results from NNWSI series 3 spent fuel dissolution tests. Pacific Northwest Laboratory, Report PNL-7170, UC-802.

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