Radiation Effects on KBS-3 Barriers
SKB's work so far

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Abstract

In the Swedish concept for final disposal of spent nuclear fuel, referred to as KBS-3, a three-layered protection system is used. The system consists of a copper canister holding the spent nuclear fuel deposited 500 meters in a repository built in groundwater saturated granitic rock. The copper canisters are placed in deposition holes, buffered and backfilled by bentonite clay. One of the challenges associated with this system is the long-term exposure of the engineered barriers i.e. the canister including the spent fuel and the cast iron insert as well as the bentonite buffer to ionizing radiation. The possible effects of radiation on the materials in the engineered barriers have been studied not only by the Swedish Nuclear Fuel and Waste Co (SKB), but also by academia nationally and internationally. In this work, literature studies have been carried out to investigate whether all possible/potential effects of ionizing radiation from the spent nuclear fuel on the engineered barriers have been considered by SKB. Apart of from a general summary of the relevant issues in chapter 1, regions within the KBS-3 design where enhanced radiation may occur have been identified and the related radiation induced processes have been summarized in chapter two. These issues include effects of water-radiolysis on the spent fuel, the cast-iron insert and other ferrous materials, the copper shell and the bentonite buffer. Three types of possible damages have been identified: the microstructural defects in the ferrous materials by direct radiation, the radiation-induced microstructural alterations of the spent fuel and the bentonite and radiation-induced oxidation i.e. dissolution of the fuel and corrosion of the ferrous metals and the copper. In chapter three, the relevant SKB documents were identified to be the SR-Site main report, the underlying Process reports and reports of FUD-programs. Apart from these documents, other SKB technical reports and open literature are used as basis for assessing whether all the issues mentioned in chapter two of this study have been considered by SKB.

According to the assessment, most of the significant issues mentioned in chapter two have been mentioned and discussed in the identified SKB documents. In the main safety assessment, i.e. the SR-Site project, the significance of most radiation processes has been based on dose levels expected at the different regions within the KBS-3 design. Irrespective of the assessments in SR-Site though, the current understanding of the relevant issues and plans for future research efforts summarized in FUD-programs reflect the research developments. However, the recent developments in understanding of radiation-induced effects on montmorillonite and the corrosion of copper in argon atmosphere need to be given due importance in coming FUD-programs. A separate investigation of radiation effects on FSW-joints may contribute to removal of the remaining uncertainties. In addition, mechanism involved in radiation-induced fuel dissolution and copper corrosion may require more research efforts than those planned.
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Introduction

Nuclear power makes up almost half of the total electricity production in Sweden. It offers a form of energy free of CO₂ emissions; however, nuclear fuel used in the reactors to generate nuclear power yields great amounts of highly radioactive nuclear waste, which requires special handling. The Swedish nuclear power industry generates 200-300 metric tons of radioactive nuclear waste annually (Nuclear Power in Sweden, 2017). There are basically two options for the handling of spent nuclear fuel namely reprocessing or final disposal. By reprocessing the spent nuclear fuel (SNF) new fuel is produced from the fissile material in the fuel. Even though this option reduces the volume of the radioactive waste, it does not totally solve the issue as there is still radioactive waste to be handled after reprocessing. In addition, there are also nonproliferation issues to be considered in terms of reprocessing as plutonium, which is used in nuclear weapons, is produced during the process. The other option, final disposal, solves the problem in terms of handling of all the waste. However, there are a great number of issues associated with this option since the final disposal of spent nuclear fuel requires long-term strategies to avoid any possible hazards from the highly radioactive waste to the human beings and the surrounding environment. (SKB Technical report P-10-47, 2010)

One of the most advanced concepts for final disposal of the spent nuclear fuel is the Swedish KBS-3 method. In this concept, a three-layered protection system, a copper canister holding the spent fuel surrounded by a buffer of bentonite clay and the surrounding natural bedrock, will prevent the escape of radioactive spent nuclear fuel to the geosphere. The spent nuclear fuel is deposited 500 meters in a repository built in groundwater saturated, granitic rock. The copper canisters are placed in deposition holes backfilled by bentonite clay (figure 1). Thus, the nuclear fuel is protected both by engineered barriers and the surrounded natural bedrock.

![Figure 1: KBS-3 method for final repository of spent nuclear fuel (source: SKB)](image)

In 2011, the Swedish Nuclear Fuel and Waste Management Company (SKB) applied to Swedish Land and Environment Court (Mark- och miljödomstolen) and Swedish Radiation Safety Authority (Strålsäkerhetsmyndigheten, SSM) for authorization to build the repository for final
disposal of Swedish spent nuclear fuel in Forsmark, Östhammar municipality, Sweden. As the projection of risks over a period as long as 100 000 years is very difficult, if not impossible, all possible present and future risks, according to SKB, are accounted for in the KBS-3 method. The three barriers are supposed to provide protection for the coming 100 000 years against risks arising from phenomena like ice age, climate change, ground water changes, falling meteorite and earth quake shifting. According to SKB, the company has been conducting scientific research for the last thirty years to ensure a safe final disposal of the nuclear fuel. One of the subjects of the research has been the effects of ionizing radiation from the spent nuclear fuel on its surroundings. (SKB, 2017) (SKB, 2006) (SKB Technical report P-10-47, 2010) (Technical report TR-10-67, 2010)

The main purpose of the project is to investigate whether all possible/potential effects of ionizing radiation from the encapsulated spent nuclear fuel within the processes that are expected to be taking place in the KBS-3 method have thoroughly been investigated. This involves identifying the processes in the KBS-3 method where ionizing radiation may occur/exist and the possibility and extent of the effects of the radiation from those processes on their respective surroundings. This is done by identifying, locating and analyzing any available literature covering issues relevant to long-term safety of deep geological repositories for nuclear fuel deposition. Examples of such literature may be Swedish and international scientific studies covering the subject, and research carried out by SKB and other stakeholders relevant for the subject.

This report contains an analysis of the research conducted thus far to address the issue as well as, based on the conducted analysis, an assessment of whether any relevant aspects related to the issue has been overlooked and need to be addressed. The study is divided into three chapters: In chapter one of the study, a general description of physico-chemical processes relevant to the long-term safety of the fuel and the engineered barriers in KBS-3 repository is given. Processes taking place within and the surrounding the fuel, the copper canister and the bentonite are shortly discussed. In chapter two, an identification of the processes in the KBS-3 concept involving enhanced levels of radiation and its possible effects on its surrounding is addressed. In chapter three, a review of the work carried out by SKB so far to address the issues discussed in chapter two i.e. the effects of the ionizing radiation, is carried out. Based on the study, an assessment of whether the issue has thoroughly been covered by SKB is made.

Delimitation

In this study, only the possible radiation processes taking place after final deposition of the canisters are addressed. Radiation issues associated with the encapsulation process i.e. the emplacement of the spent fuel into the copper canister and those with the transport of the spent fuel from the encapsulation plant to the repository site are not considered.
ABBREVIATIONS

bcc - body centered cubic
BWR - Boiling-water react
CEC - Cation Exchange Capacity
CLAB - Centralt mellanlager för använt kärnbränsle
Cu-OF - Oxygen-free copper
dpa - Displacements per atom
fcc - face-centered cubic
FSW - Friction Stir Welding
FUD - Program för forskning, utveckling och demonstration av metoder för hantering och slutförvaring av kärnavfall
HLW - High-level waste
KBS-3 - KärnBränsleSäkerhet (the Swedish spent fuel repository design)
KTH - Kungliga Tekniska Högskolan
MOx - Mixed oxide (fuel)
NDT - Non-Destructive Testing
NPP - Nuclear Power Plant
PWR - Pressurized-water reactor
R&D - Research and Development
sc - simple cubic
SCC - Stress Corrosion Cracking
SFR - Slutförvaring av radioaktivt avfall
SKB - Svensk Kärnbränslehantering
SNF - Spent Nuclear Fuel
SP - Stopping power
SRB - Sulphate Reducing Bacteria
SR-Site - The Safety Assessment Project
SSM - Strålsäkerhetsmyndigheten
Chapter One
General summary of the issues

In this chapter, an overview of issues involving the spent nuclear fuel and the surrounding engineered barriers in the deep geological repository is given. Processes directly involving the spent fuel, such as the long-term evolution of the fuel in the repository atmosphere, dissolution of UO$_2$ and corrosion of the cladding, are discussed under section 1 (The spent nuclear fuel). This is followed by discussions of issues relating to the copper canister and the bentonite barrier under section 2 and 3, respectively.

1 The spent nuclear fuel

The fuel used in nuclear reactors to generate electricity consists of almost stoichiometric uranium dioxide in the form of cylindrical pellets with varying sizes but in the order of 1 cm in length and 1 cm in diameter. The pellets are stacked in about 4-meter-long zirconium alloy tubes referred to as fuel cladding. These tubes are bundled into fuel assemblies (figure 2). Apart from zirconium alloy, the structural material of fuel assembly contains nickel alloys Inconel and incoloy as well as stainless steel. (Ewing, 2015)

![Figure 2: Zircalloy tube (cladding) containing cylindrical UO$_2$ pellets. (b) Fuel assembly (Technical report TR-10-46, 2010)](image)

The natural occurring concentration isotope $^{235}$U, the fissile material in the nuclear fuel, is 0.7%. The UO$_2$ in the fuel is enriched in isotope $^{235}$U to about 3.6% for BWR fuel and 4.2 % in PWR fuel. This enrichment level is planned to be increased in the future. In order to generate energy,
the fuel is irradiated by neutrons in the reactor leading to fission of U (nuclear chain reaction) and release of 200 MeV of energy per nucleus of U (Technical report TR-10-46, 2010) (Ewing, 2015)

1.1 Characteristics of the spent nuclear fuel
When the nuclear fuel has been used up and taken out of the reactor, it is called spent nuclear fuel (SNF). Due to very high levels of radioactivity, SNF needs special handling. The composition of the SNF is determined mainly by the following two reactions in the reactor: Fissile nuclide fission, such as fission of \(^{235}\text{U}\) and \(^{239}\text{Pu}\) and neutron capture followed by beta decay reaction leading to production of transuranium isotopes, mainly \(^{238}\text{U}\) and \(^{239}\text{Pu}\). Even though several hundred different fission products are formed in the reactor, most of them decay away due to their short half-lives. The final composition depends on factors such as type of the fuel, enrichment level and chemical composition of the fuel used in the reactor, neutron energy spectrum and burn-up of the fuel. The burn-up are typically in the range of 35 – 45 MWd/kgU. However, nuclear power plant operators tend to desire higher burn-ups in order to extract more energy from the nuclear fuel. This is acquired through different means such as control of water chemistry in the reactor. As a rule of thumb, a burn-up level of 30 MWd/kgU is expected to yield a conversion of four atomic percent of the uranium to about three atomic percent fission products and 1% transuranium isotopes. (Ewing, 2015) (Choppin, Liljenzin, Rydberg, & Ekberg, 2013)

It is rather the effects of the individually released radionuclides that determine the radiotoxicological impacts in performance assessment of repositories for long-term SNF deposition than the amount of released species. To be able to make assessments about the long-term evolution of SNF, it is therefore important to understand the amounts, spatial distribution and radioactivity of the radionuclides. (Carbon, Wegen, & Wiss, 2015)

SNF mainly consists of UO\(_2\) (96 %), mostly \(^{238}\text{U}\), but also some unfissiated \(^{235}\text{U}\). As \(^{238}\text{U}\) has very long half-life of 4.699×10\(^9\) years, its concentration remains constant even after 10\(^6\) year. \(^{238}\text{U}\) converts to \(^{239}\text{Pu}\) through neutron capture reactions and two beta decays. \(^{239}\text{Pu}\) may also undergo neutron capture and form \(^{240}\text{Pu}\). As generation of the both isotopes of Pu grow during radiation, it is the element with the second highest concentration in SNF. (Carbon, Wegen, & Wiss, 2015)

The remaining portion, fission products, transuranium isotopes and activation products, occur in different phases and structural forms. Gases in the fission products, such as I, Xe and I, form finely dispersed bubbles in the fuel grains. (Werme, Lilja, Sellin, & Spahić, 2010). Metallic fission products, for example Ru, Rh, Mo, Tc and Pd, form immiscible metallic precipitates, so called \(\varepsilon\)-particles, ranging in sizes from nanometers to micrometers. Some fission products form oxide precipitates of Zr, Rb, Cs, Ba and others, for example Sr, Zr, Nb and lanthanides, may form solid solutions with the UO\(_2\). In addition, some transuranium elements may substitute for U in UO\(_2\) (figure 3). Due to very sharp thermal gradients with temperatures as high as 1,700°C at the center of the pellet and declining to 400 °C at its rim, the element distribution is not even homogenous within a single pellet. (Ewing, 2015) (Technical report TR-10-46, 2010) (Carbon, Wegen, & Wiss, 2015)
In Sweden, light water reactors, both of boiling water (BWR) and pressurized water (PWR) types, are used for power generation. The KBS-3 repository will receive spent nuclear fuel mostly in the form of UO$_2$ from the operating PWR and BWR reactors in Sweden. However, some quantities of other fuel types, such as MOX fuel, spent fuel from heavy water reactor of Ågesta and fuel residues from Studsvik will also be included in the fuel types that will be deposited in the final repository. The average burnup of Swedish BWR fuel is 34 MWd/kgU and that of PWR is 41 MWd/kgU according to an assessment by SKB (Technical report TR-10-46, 2010).

There are differences in the level of burn-up across the UO$_2$ pellet. The edges of the pellet have higher burn-up which leads to higher concentrations of $^{239}$Pu as well as more porous structure and smaller grain size at these spots. Thermal excursions caused by steep temperature gradients during reactor operation may cause structural alteration of the fuel such as coarsening of the grain size and extensive microfracturing. Some volatile species, for example Cs and I, may migrate to different spots within the fuel microstructure such as grain boundaries, fractures as well as the gap between fuel pellet and the zircalloy tube cladding (figure 3). Understanding the distribution of the radionuclides within the fuel microstructure is important since the fuel that migrate to different spots within the microstructure of the UO$_2$ may release faster when fuel is exposed to water than those in the fuel matrix. (Ewing, 2015) (Guenther, Blahnik, Thomas, Baldwin, & Mendel, 1990) (Ball, Burns, Henshaw, Mignaneli, & Potter, 1989). Spent nuclear fuel has a very complex chemistry and phase distribution. Understanding these factors are important to assess the long-term changes of the spent fuel in the repository environment. (Ewing, 2015)
1.2 Long-term radioactivity

The radioactivity of the spent nuclear fuel changes with the passage of time. Initially, most of the radioactivity originates from short lived fission products. The main contributors of radioactivity will change from the initial short-lived fission products such as $^{137}\text{Cs}$ and $^{90}\text{Sr}$ (half-lives of about 30 years) to much longer-lived actinides such as $^{99}\text{Tc}$ (half-life of 210000 years), $^{79}\text{Se}$ (1.1 million years), $^{135}\text{Cs}$ (2.3 million years) and $^{129}\text{I}$ (16 million years). Over even longer timescales, actinides such as $^{238}\text{U}$ (with a half-life of 4.5 billion years), $^{235}\text{U}$ (0.70 billion years) and $^{237}\text{Np}$ will become the main contributors of activity (Carbon, Wegen, & Wiss, 2015) (Ewing, 2015).

Initially the spent fuel has a very high level of radioactivity, about 100000 times more than unirradiated fuel (10 GBq/MT of fuel at the time of removal from the reactor). The thermal output is as high as 2MW/MT, which is only 10% of the in-reactor thermal output. Within a hundred years the thermal output decreases to less than 1% of its initial value. (Ewing, 2015) (Carbon, Wegen, & Wiss, 2015)

The decay of short lived radionuclides such as $^{137}\text{Cs}$-$^{137m}\text{Ba}$, $^{89}\text{Sr}$-$^{90}\text{Y}$, $^{91}\text{Y}$, $^{95}\text{Zr}$-$^{95}\text{Nb}$, $^{85}\text{K}$ and $^{106}\text{Ru}$-$^{106}\text{Rh}$ emit a very intense $\beta(\gamma)$-field when the SNF is fresh. Within the following 300-500 years in the repository, however, most of the nuclides emitting $\beta(\gamma)$ radiation will have decayed and $\alpha$-radiation will be the dominating type of radiation (figure 4). The remaining $\beta$-activity after 1000 years is due to fission products with longer half-lives, such as $^{79}\text{Se}$, $^{92}\text{Zr}$, $^{99}\text{Tc}$, $^{129}\text{I}$, $^{94}\text{Nb}$, $^{135}\text{Cs}$ and decay chains of $^{233,236,238}\text{U}$ emitting $\beta$-decay.

![Figure 4: $\alpha$ and $\beta$ radioactivity in SNF with a burn-up of 40Gwd/THM (tons heavy metal) as a function of cooling time. The dotted line represents alpha activity before irradiation in a PWR.](Carbon, Wegen, & Wiss, 2015)
The decay of $^{242,244}$Cm, $^{238}$Pu and $^{241}$Am are the main sources of the initial $\alpha$-activity while the late decrease is mainly caused by the decay of long-lived sources of $\alpha$-radiation such as $^{239,240}$Pu and $^{243}$Am. (Carbon, Wegen, & Wiss, 2015)

1.3 Long term stability of SNF
There are several issues in terms of long term mechanical and chemical stability of SNF that need to be considered. These issues are summarized in the following lines:

1.4 Mechanical stability
Some issues affecting the mechanical properties of the SNF are the following: Microstructural changes in SNF, helium gas build up, grain disintegration, dislocations, hardness, heat capacity and swelling. SNF stability, in case of no water-intrusion, is mainly determined by radiation damage, helium build-up and variation in oxygen potential. In case of contact with water, a possible preferential dissolution of grain boundaries, rather than the matrix, will lead to an increase in the fuel wet surface area. This will cause parts of the inventory to become available for fast release and mobilization. Apart from radionuclide release, the contact with water will lead to weakening of cohesion between UO$_2$ grains causing a general deterioration of the mechanical stability of the fuel. In addition, the increased wetted surface area will lead to more oxidants produced through radiolysis of water by $\alpha$-radiation. (Marchetti, Belloni, Himbert, Carbol, & Fanghänel, 2010) (Carbon, Wegen, & Wiss, 2015)

As far as radiation damage is concerned, it has been observed that more plutonium is built up at the edges (“rim” region) of the fuel than in the central parts during irradiation in the reactor due to neutron resonant capture cross-section of $^{238}$U. Owing to the characteristic neutron energy spectrum in light water reactors, a higher density of epithermal neutron resonance absorption in $^{238}$U nuclei exists at the edges of the fuel pellet as compared to the central part of the fuel. This causes a local enrichment in $^{239}$Pu through $\beta$-decay of $^{239}$Np resulting in a locally higher fission density in the rim-region. The higher burn-up of plutonium in this region leads to creation of a structure referred to as High Burnup Structure (HBS) characterized by dense small subgrains about 200 nm in size and accumulation of small pores about 1µm in size. Apart from effecting the fuel performance in terms of fission gas release, the temperature of the fuel, its hardness and swelling, HBS is the first exposed layer in case of contact with water. (Fors, Winckel, & Spahiu, 2009) (Carbon, Wegen, & Wiss, 2015)

In addition, it has been observed that $\alpha$-radiation displaces atoms in the lattice leading to an increase in lattice parameter resulting in microscopic swelling. (Xiao, Long, & Hongsheng, 2016) (Carbon, Wegen, & Wiss, 2015)

In light water reactor fuel, both UO$_2$ and MOX, significant amounts of helium are produced by $\alpha$-decay of fissile atoms and heavier nuclides produced by neutron capture. One of the main issues concerning alpha decay is the low solubility of helium in the UO$_2$ lattice resulting in its precipitation in the spent fuel matrix. This may cause microscopic and macroscopic swelling of SNF leading a deterioration of the mechanical properties of the fuel. The release of helium gas in SNF may also lead to an internal higher pressure on the cladding, which may cause rupture of the
cladding which functions as the first barrier preventing the release of radionuclides from SNF. (Talip, o.a., 2013) (Carbon, Wegen, & Wiss, 2015) (Ewing, 2015) (Talip, o.a., 2013) (Ronchi & Hiernaut, 2004)

Since 1960s, studies have been carried out to measure diffusion coefficient in UO$_2$. However, there have been considerable discrepancies in the results obtained by different studies (Carbon, Wegen, & Wiss, 2015). Some studies have suggested that diffusion of helium within UO$_2$ is negligible considering the repository conditions and time scales (Roudil, o.a., 2003) while others (Talip, o.a., 2013) (Ronchi & Hiernaut, 2004) (Talip, o.a., 2013) have observed opening of grain boundaries caused by $\alpha$-radiation damage leading to release of significant amounts of radiogenic helium, through diffusion accumulated in the fuel samples.

1.5 Chemical Stability
The chemical stability of the SNF depends on a number of issues, which are generally discussed in the lines below.

1.5.1 Fuel dissolution
The dissolution of UO$_2$, in case of contact with water, will be determined by the solubility of the UO$_2$ in groundwater. The solubility will depend on groundwater chemical composition and factors such as pH, temperature, ionic strength, complexing ions as well as crystallinity of UO$_2$. The perfect structure of fluorite lattice formed by U and O atoms in crystalline UO$_2$ (cr) makes it less susceptible to dissolution in water since this will require breakage of many bonds. Surface U(IV) atoms in contact with water, however, are strongly hydrated which facilitates breakage of the bonds since the coordination of water molecules takes place already on the UO$_2$ surface before detachment (Langmuir, 1978) (Carbon, Wegen, & Wiss, 2015)

The fuel pellet could be divided in three regions in terms of radionuclide release in case of contact with water: 1- Fuel-cladding gap, crack surfaces and open porosity; 2 – grain boundaries; 3 – The UO$_2$ matrix (figure 5). Upon contact with water i.e. in case of breach of the fuel cladding and other engineered barriers, the radionuclides in the fuel-cladding gap will be the first to release and dissolve in the water. The subsequent dissolution rate of radionuclides from the grain boundaries will be higher than those from the fuel matrix. However, in the long-term the rate of nuclide release is thought to be governed by dissolution of the UO$_2$ matrix. (Carbon, Wegen, & Wiss, 2015) (Casella, Hanson, & Miller, 2016) (Shoesmith D., 2000)
1.5.2 Importance of redox conditions

One of the characteristics of oxide dissolution processes is the occurrence of a wide range of dissolution rates varying by order of magnitude between different oxides and even the same oxide. $\text{UO}_2$ has been categorized as slowly dissolving semiconductors. (Nowotny & Dufour, 1988)

For this category of oxides, the processes that control the rate of dissolution is either charge transfer to the surface for formation of surface ionic species ($M^{n+}, O^{2-}$) which can then transfer to solution or surface alterations that lead to formation of these transferable ionic species. Therefore, the kinetics of dissolution is governed mainly by properties such as redox potential of the solution, solid state conductivity, and ion formation at surface defect sites. As far as $\text{UO}_2$ is concerned, the redox potential is of utmost importance since oxidation of $\text{UO}_2$ (to $\text{UO}_2^{2+}$)
increases its solubility in water by several orders of magnitude. Thus, the redox conditions in the repository control the solubility of the spent fuel. (Shoesmith D., 2000)

The environment in the deep geological repository is characterized by reducing conditions due to absence of dissolved atmospheric oxygen and other oxidizing agents. The reducing conditions may however be altered by formation of oxidizing species such as H₂O₂ by radiolysis of water close to the surface of the fuel caused by α-radiation from the SNF. The oxidizing radiolysis products may oxidize UO₂ to UO₂⁺ making it a 1000 times more soluble in water. The higher the burn-up of the fuel, the higher the extent of α-radiolysis will be, which means more oxidants will be available to oxidize UO₂. Spent fuel with a normal burn-up level that has been stored for 3000 years will emit 1000 α particles/mm², s, which in turn will generate 5×10⁴ molecules of H₂O₂. Considering the repository time scales, there will be enough oxidizing species to completely oxidize the fuel. However, there have been studies which show that this tendency will be countered by large amounts of H₂ produced through anoxic corrosion of iron in (figure 7). (Carbol, Fors, Thomas, & Spahiu, 2009)(Bruno & Spahiu, 2014) (Carbon, Wegen, & Wiss, 2015) (Shoesmith D., 2000)

![Figure 7](image)

**Figure 7: Schematic illustration of fuel dissolution by water radiolysis products.** (Shoesmith D., 2000)

In addition, the effects of the metallic inclusions, the so called ε-particles, have been studied which have an influence over the UO₂ dissolution rate (Carbol, Fors, Thomas, & Spahiu, 2009) 2009) (Carbon, Wegen, & Wiss, 2015) (Nowotny & Dufour, 1988) (Jonsson, 2012) (Shoesmith D., 2000) (Wu, Qin, & Shoesmith, 2014). These issues and other radiation induced processes are discussed in chapter two and chapter three of this study.

1.6 Effects of groundwater composition

The composition of the groundwater will change in respect to the composition of bulk water in contact with the bedrock as it passes through the different engineered barriers, in case of failure of these barriers, towards the fuel. This is because water passing through the barriers will
incorporate species in the barriers such as those in the bentonite clay and the fuel cladding. The composition of ground water in Forsmark site and Äspö are compared in table 1. The difference in composition of granitic groundwater in Äspö, Sweden and that of the same groundwater equilibrated with bentonite (MX-80) can be seen in the table. Concentrations of ions such as Na, K, Ca order of magnitude higher in groundwater equilibrated with bentonite. The dissolution of calcite (CaCO$_3$) impurities present bentonite release carbonate ions; higher concentrations of carbonate lead to lowering of pH in the groundwater. Additionally, sulfate concentration in the groundwater increases due to the presence of CaSO$_4$ in bentonite. (Carbon, Wegen, & Wiss, 2015) (Hunter, Fiona, Tim, & Hoch, 2007)

The difference between groundwater composition in Forsmark site and those of the Äspö depends on the fracture pathways and orientation and the fracture filling minerals the groundwater has exchanged minerals with. Considering the repository timescales, groundwater composition may also change due to processes occurring during glaciations and deglaciations. (Björk & Svensson, 1992) (Carbon, Wegen, & Wiss, 2015) (Casella, Hanson, & Miller, 2016)

Table 1: Variation in groundwater composition in different sites considered for long-term geological repository (Carbon, Wegen, & Wiss, 2015) (Guimerà, Duro, Delos, & Spain, 2006) (Metz, Kienzler, & Schußler, 2003) (Hunter, Fiona, Tim, & Hoch, 2007) (Björk & Svensson, 1992)

<table>
<thead>
<tr>
<th>Components</th>
<th>Granitic groundwater at Forsmark, Sweden (mol kg$^{-1}$)</th>
<th>Granitic groundwater at Äspö, Sweden (mol kg$^{-1}$)</th>
<th>Granitic groundwater from Äspö equilibrated with bentonite (mol kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na$^+$</td>
<td>$8.9 \times 10^{-2}$</td>
<td>$9.1 \times 10^{-2}$</td>
<td>$6.4 \times 10^{-1}$</td>
</tr>
<tr>
<td>K$^+$</td>
<td>$8.8 \times 10^{-4}$</td>
<td>$2.1 \times 10^{-4}$</td>
<td>$3.1 \times 10^{-3}$</td>
</tr>
<tr>
<td>Ca$^{2+}$</td>
<td>$2.3 \times 10^{-2}$</td>
<td>$9.6 \times 10^{-3}$</td>
<td>$1.0 \times 10^{-2}$</td>
</tr>
<tr>
<td>Mg$^{2+}$</td>
<td>$9.3 \times 10^{-3}$</td>
<td>$1.7 \times 10^{-3}$</td>
<td>$4.9 \times 10^{-3}$</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>$3.3 \times 10^{-5}$</td>
<td>$4.4 \times 10^{-6}$</td>
<td>$2.5 \times 10^{-4}$</td>
</tr>
<tr>
<td>Fe$^{3+}$</td>
<td>$1.8 \times 10^{-4}$</td>
<td>$1.5 \times 10^{-4}$</td>
<td>$3.2 \times 10^{-4}$</td>
</tr>
<tr>
<td>Mn$^{2+}$</td>
<td>$1.8 \times 10^{-3}$</td>
<td>$7.9 \times 10^{-5}$</td>
<td>$6.4 \times 10^{-1}$</td>
</tr>
<tr>
<td>Cl$^-$</td>
<td>$1.8 \times 10^{-1}$</td>
<td>$1.8 \times 10^{-1}$</td>
<td>$1.6 \times 10^{-2}$</td>
</tr>
<tr>
<td>Br$^-$</td>
<td>$3.0 \times 10^{-3}$</td>
<td>$5.0 \times 10^{-4}$</td>
<td>$2.5 \times 10^{-3}$</td>
</tr>
<tr>
<td>HCO$_3^-$</td>
<td>$1.8 \times 10^{-3}$</td>
<td>$1.6 \times 10^{-4}$</td>
<td>$1.6 \times 10^{-2}$</td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>$6.8 \times 10^{-3}$</td>
<td>$5.8 \times 10^{-3}$</td>
<td>$1.6 \times 10^{-2}$</td>
</tr>
<tr>
<td>T ($^\circ$C)</td>
<td>15</td>
<td>20</td>
<td>Oxidizing</td>
</tr>
<tr>
<td>kg pCO$_2$</td>
<td>7.0</td>
<td>8.1</td>
<td>8.4</td>
</tr>
<tr>
<td>pH</td>
<td>-0.143</td>
<td>-0.258</td>
<td></td>
</tr>
</tbody>
</table>

The composition of water is important for the UO$_2$ dissolution in case of water contact with the SNF. The rate of dissolution will depend on concentrations of different ions in the groundwater and their respective tendency to build ionic complexes since the solubility of different ionic complexes will control when saturation is reached. High concentration of U(VI) dissolved in
groundwater together with alkali metals may lead to formation of precipitates such as sodium diuranate (Na₂O (UO₂)₂·6H₂O). In the presence of Si and alkaline metals, such as Ca, Mg, U(VI) may form silicates such as uranophane, Ca(UO₂)₂(SiO₃OH)₂·5H₂O. These U(VI) containing silicate minerals keep the U(VI) concentration below 10⁻⁶ M due to their relatively low solubility in the presence of silicates. Another U(IV)-containing silicate mineral, USiO₄ (cr) (Coffinite), which is primarily found in association with UO₂(cr) and SiO₂ (cr) is believed to have lower solubility than UO₂(cr) in the presence of high concentrations of silica. (Guo, o.a., 2015) (Grenthe, o.a., 1993) (Carbon, Wegen, & Wiss, 2015) (Shvareva, o.a., 2011)

Reducing conditions prevail the groundwater, as evident from the presence of unoxidized siderite and pyrite, in the granitic deep geological repositories due to the absence of dissolved atmospheric oxygen. Iron and manganese occur in their reduced forms and influence the redox potential of the groundwater. Considering these facts, a redox potential below zero is expected in the groundwater. As for the anions in the groundwater, Br and Cl build weaker complexes with cations and U(IV) as compared to OH⁻ and CO₃²⁻. Cl and Br may, however, cause the redox conditions to move towards oxidizing by scavenging radiolysis products such as OH-radicals and limiting the production of H-radical. (Carbon, Wegen, & Wiss, 2015)

1.7 Cladding degradation
Zirconium alloys (Zircalloy) are widely used as fuel cladding and structural material in nuclear reactors due to their corrosion-resistant properties and low neutron absorption cross section. The chemical composition of zirconium alloys, Zircaloy-2 (Zr-2) and Zircaloy-4 (Zr-4), used as fuel cladding in the nuclear reactors is given in table 2.

Table 2: Composition (in mass %) of Zircalloys used in nuclear reactors according to ASTM specifications (Shoesmith & Zagidulin, 2011)

<table>
<thead>
<tr>
<th>Alloying elements</th>
<th>Zircaloy-2</th>
<th>Zircaloy-4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sn</td>
<td>1.2–1.7</td>
<td>1.2–1.7</td>
</tr>
<tr>
<td>Fe</td>
<td>0.07–0.20</td>
<td>0.18–0.24</td>
</tr>
<tr>
<td>Cr</td>
<td>0.05–0.15</td>
<td>0.07–0.13</td>
</tr>
<tr>
<td>Ni</td>
<td>0.03–0.08</td>
<td>–</td>
</tr>
<tr>
<td>Nb</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

As seen in the table, Sn which is an alpha-phase stabilizer is the primary alloying component added to the alloys to improve their corrosion resistance properties. Iron, chrome and nickel are added in small amounts for the enhanced corrosion resistance and mechanical strength. In Zr-4 higher Fe-iron content and no addition of Ni as compared to Zr-2 can be observed. As Nickel may function as catalyst for absorption of H, its addition is avoided in Zr-4. (Shoesmith & Zagidulin, 2011)

Due to exposure to irradiation during reactor operation, neutron activation products, such as ⁵⁹Ni, ⁶³Ni, ¹⁴C and ⁹³Zr is formed in fuel cladding. The release of these radionuclides due to corrosion
should be considered as part of long-term safety assessment of deep geological repositories. (Shoesmith & Zagidulin, 2011)

Zr undergoes corrosion to build a relatively thick surface oxide (about 4 µm) during contact with water under in-reactor irradiation:

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$$

During the process, a considerable amount of hydrogen is absorbed into the alloy microstructure. An unirradiated Zr-alloy cladding has a residual content of 25µg/g of hydrogen which increases to 100 µg/g during reactor operation. When removed from the reactor, hydrogen may precipitate as zirconium hydride during the cooling process of the cladding, which may lead to embrittlement and an increased risk of fracture (Shoesmith & Zagidulin, 2011).

Zr-alloys contain alloying elements and impurities with relatively low solubility in α-Zr matrix. These insoluble species migrate to grain boundaries and form intermetallic precipitates such as Zr₂(Fe, Ni) and Zr (Fe, Cr)₂. In the redox conditions expected in the repository, this may not have any major influence on the passivity of the oxide.

In case of water intrusion through the engineered barriers, the corrosion of zircalloy cladding will depend on groundwater composition, and other properties such as its redox potential and pH. There are mainly two processes which could lead to corrosion of Zr alloys namely the breakdown of the passive film and subsequent pitting corrosion and absorption of hydrogen leading to embrittlement under anoxic reducing conditions. In figure 8, a schematic illustration of potential ranges under which passive film break down and absorption of hydrogen may occur. The corrosion potentials ($E_{corr}$) were measured in a series of solutions with neutral pH, and sulfate chloride or perchlorate in 0.1 mol/L concentrations. The positive potential limit shows the potential values acquired in aerated solutions and those with negative limit were obtained under Ar-purged conditions. (Shoesmith & Zagidulin, 2011)

![Figure 8: Schematic illustration of potential ranges under which passive film break down and hydrogen absorption may occur. (Shoesmith & Zagidulin, 2011)](image)
The presence of Cl\(^-\) in the groundwater may pose a risk of the oxide film breakdown and pitting corrosion if the conditions are oxidizing enough to polarize the \(E_{corr}\) to the potential range (figure 8) under which such a process may take place. As reducing conditions prevail the groundwater in deep geological repositories, the only oxidants that may alter the redox conditions are the ones produced by radiolysis of water by radiation from SNF. On the inner surface of the cladding, due to short range of \(\alpha\)-radiolysis, \(H_2O_2\) will be the main oxidant while gamma radiolysis products will be formed on the outer surface of the cladding. For the negative potentials to be enough for the hydrogen embrittlement, the corrosion potential (\(E_{corr}\)), must be polarized to more negative values (Shoesmith & Zagidulin, 2011). The alteration of the groundwater redox conditions due to radiolysis of water will be in more details in chapter two.
2 The copper canister

The canister is supposed to hold the fuel assemblies containing the fuel rods with spent nuclear fuel. The canister will be placed in deposition holes surrounded by a buffer of bentonite clay whose functions are described under a separate section (section 3) of this chapter. The main function of the canister is to protect the fuel and isolated it from the surrounding environment. In the KBS-method, copper has been chosen to be the material for the canister (the canister tube, lid and base of the canister) due to its natural corrosion-resistant properties. The fuel assemblies are placed in the insert made of iron cast in the copper canister (figure 1). The mechanical properties of the insert are important in terms of resistance to mechanical loads, brittleness due to radiation and fracture strength (Technical report TR-10-67, An update of the state-of-the-art report on the corrosion of copper under expected conditions in a deep geologic repository, 2010).

Figure 9: The copper canister and the insert (Technical report TR-10-67, 2010)

High purity, oxygen free copper (Cu-OF) is used as the construction material for the canister ensuring properties such as high creep ductility and corrosion resistance. Phosphorous (100 ppm) is added to enhance the creep strength and the creep ductility (Technical report TR-10-46, 2010; Technical report TR-10-67, 2010).

The canister itself is a hot extruded cylindrical tube with a lid and a bottom forged by a solid-state joining technique FSW (friction stir welding). The issue of defect formation often experienced in the conventional fusion welding is avoided by applying FSW. This technique is described as a lower energy input process as compared conventional fusion welding. However, the heat produced during the welding process is high enough to cause sharp thermal gradients and localized expansions in the copper pieces being forged together by the technique. During the cooling process, some areas may contract more as compared to other areas in the copper pieces forged together. This may lead to misfits between different areas giving rise to permanent strains,
which may consequently result in residual stress. As alloyed copper is susceptible to SCC (stress corrosion cracking) under certain conditions, the residual stress being one contributing factor, the importance of keeping the residual stress induced during the manufacturing process as low as possible. (Jin & Sandström, 2012) (Technical report TR-10-67, 2010)

The long-term resistance of the copper canister to corrosion is, apart from the copper’s good corrosion-resistant properties, based on the fact that the bentonite, highly compacted after absorbing water, will be firmly in contact with the copper canister leading to absence or lack of oxygen very close to the copper surface, thus leading to a decreased possibility for oxidation (corrosion) of the copper material. In addition to preventing the transport of contaminants to the copper surface, the close contact of the bentonite buffer with the canister is supposed to be helpful in avoiding bacterial activity between the canister/bentonite interface, which may help in avoiding corrosion due to sulfide production by sulphate reducing bacteria (SRB). (Technical report TR-10-67, 2010)

The long-term capability of copper to withstand corrosion has been studied over the past years and the notion of its long-term stability against corrosion has been questioned. The issues related to the possibility of copper corrosion are generally discussed in the lines below.

2.1 Copper canister corrosion
During the initial period after the deposition of the copper canisters, the Cu corrosion will depend on the factors such as moisture, temperature and the existence and concentrations of other reactive species such as chlorides, sulphates and nitrates. The changes in the repository environment in terms of redox conditions and changes in groundwater chemistry over in long-term perspective will eventually lead to steady state conditions. The long-term corrosion of Cu will therefore depend on the surrounding environment (pH, chemical composition of ground water, resistivity). (King, Kolar, Vähänen, & Lilja, 2011)

The initial oxygen content will be consumed by reactions with the minerals and organic matter in the bedrock and bentonite clay as well as by initial corrosion of the copper canister. In the long-term perspective, i.e. when the trapped oxygen has been consumed, the corrosion of the copper canister will depend on the availability HS− at the surface of the copper canister. (King, Kolar, Vähänen, & Lilja, 2011) (Chen, Qin, & Shoesmith, 2011).

There have, however, been studies which claim that water can oxidize copper under certain conditions relevant for the deep geological repositories (Hultquist*, 1986) (Hultquist, o.a., 2015). In addition, the redox conditions in the repository environment can be altered by the radiolytically produced oxidants (Jonsson, 2012). These issues are discussed under respective sections in the lines below.

2.2 Sulfide induced corrosion
According to King et al. (King, Kolar, Vähänen, & Lilja, 2011), the long-term corrosion of the copper canister in the repository environment will only be determined by the availability of sulfide ions at the surface of the copper canister after the oxygen initially trapped in bentonite
barrier/backfill and any Cu(II) formed due to homogeneous oxidation of Cu(I) by the initially trapped oxygen have been consumed. (King, Kolar, Vähänen, & Lilja, 2011)

Sulphide in the deep ground waters may be originating from dissolution of sulphide containing minerals, reduction of sulphate by SRB (sulphate reducing bacteria) or dissolved sulfide in the groundwater. It has been argued that the origin of the dissolved sulfide may be pyrite (FeS₂); however there has not been enough evidence of FeS₂ dissolution in anaerobic conditions prevailing in the deep groundwater. The possibility of reduction of sulphate by microbial activity at the canister/bentonite surface may be very limited due to the high pressure of bentonite compaction. However, sulfide produced by SRB in the far field can diffuse through the bentonite to the surface of the copper canister. Surface corrosion by sulfide diffused through the bentonite clay buffer to the surface of the copper canister is estimated to be in the order of nanometers per year by Swedish Nuclear Fuel and Waste Management Company. (King, Kolar, Vähänen, & Lilja, 2011) (SKB, 2006) (King*, Lilja, & Vähänen, 2013)

Studies investigating adsorption of ions on Cu surface and the corrosion of copper by sulfide have found that the corrosion process may start with adsorption of SH⁻ on the surface of the copper:

\[ Cu + SH^- \rightarrow Cu(SH)_{ads} + e^- \]

This may be followed by the production of Cu₂S in a reaction step involving the adsorbed Cu(SH)_{ads} and Cu producing Cu₂S:

\[ Cu + Cu(SH)_{ads} + SH^- \rightarrow Cu_2S + H_2S + e^- \]

Copper sulfide (Cu₂S) film was observed to form in the long-term experiments carried out by Chen et al. (Chen, Qin, & Shoesmith, 2011). Cu₂S was found to grow at constant rate after the formation of Cu₂S covered the Cu surface, indicating that the film may be only partially protective and Cl- may, through complexation and solubilization of Cu(I), facilitate the corrosion process:

\[ Cu(SH)_{ads} + 2Cl^- \rightarrow CuCl_2^- + SH^- \]

Due to low solubility of Cu(I) in SH⁻ containing solution, the dissolved Cu(I) may precipitate as Cu₂S (Chen*, Qin, & Shoesmith, 2010):

\[ 2CuCl_2^- + SH^- \rightarrow Cu_2S + 4Cl^- + H^+ \]

Studies of the kinetics of the formation of Cu₂S on the surface of copper by Chen et al. (Chen*, Qin, & Shoesmith, 2010) show that depending on the concentration of SH-, Cu₂S layer may demonstrate different growth behavior. In case of higher concentrations (5.0 × 10⁻⁴ M), it was found by Chen et al. that the growth of Cu₂O film follows a parabolic law and is controlled by transport of the Cu⁺ in the microstructure of Cu₂S film, either through Cu₂S matrix or along the grain boundaries. In a study by the same authors, it was observed that in case of a lower concentration of HS⁻ (5.0 × 10⁻⁵ M), film growth follows a linear growth law and is governed by
diffusion of SH\textsuperscript{+} ions yielding a film with a porous and non-protective structure. (Chen**, Qin, & Shoesmith, 2011).

In a study carried out by Taylor et al. (Taylor, Partovi-Nia, Chen, Qin, & Shoesmith, 2014), the film growth was studied in solutions with a range of SH\textsuperscript{+} and Cl\textsuperscript{−} concentrations to investigate the passivation capability of the film and possible breakdown of the film leading to pitting corrosion of copper. Formation of three different surface layers of Cu\textsubscript{2}S were observed: a single porous layer (referred to as Type I), a double layer film (Type II) and a rather compact film (Type III), offering partial passivation. It was observed that the Type I and II surface films are formed when the film growth is governed by diffusion of sulphide in the solution while Type III is formed when the interfacial reactions are the controlling factor of the growth of the film. For the Type III films to form, high SH\textsuperscript{+} concentrations and high flux of sulphide ions at the film/electrolyte interface are required. This means, according to the authors, that the possibility of pitting corrosion is minimal since the conditions necessary for formation of the Type III (partially passive) films, cannot be expected in the deep geological repositories. (Taylor, Partovi-Nia, Chen, Qin, & Shoesmith, 2014)

2.3 Corrosion in absence of sulfide

In a study by Hultquist (Hultquist*, 1986) more than 30 ago, it was claimed that Cu can be oxidized by H\textsubscript{2}O with evolution of H\textsubscript{2} i.e. corrosion of Cu in absence of sulfide or dissolved oxygen may be possible. Hultquist and co-workers have through a series of studies since then made several observations supporting the claim. There have also been a number of studies, mostly by King et al. arguing against the claims made by Hultquist and co-workers. (Lilja & King, 2011).

The claims made by Hultquist differ fundamentally from the general conception of Cu corrosion. The classic thermodynamic approach to Cu corrosion, as can be seen in the pourbiax diagram for Cu-H\textsubscript{2}O (figure 2), has been that under standard temperature and pressure (25°C and 1 atm) the only stable solid copper products formed are Cu, Cu\textsubscript{2}O or CuO.
This means that the reaction of Cu with water (the reaction below) and the evolution of H$_2$ from reduction of H$_2$O at atmospheric pressure differ in equilibrium potentials by 470 mV:

$$2Cu + H_2O \rightarrow Cu_2O + 2H^+ + 2e^-$$

Thus, for oxidation of Cu by H$_2$O (the following reaction) to occur spontaneously, a partial pressure of 10$^{-16}$ atm of H$_2$ is required.

$$2Cu + H_2O \rightarrow Cu_2O + H_2$$

Hultquist and co-workers explain that the corrosion of Cu is caused by formation of the previously unknown phase of copper H$_x$CuO$_y$. The authors argue that H$_x$CuO$_y$ is a thermodynamically stable phase since the equilibrium partial pressure of H$_2$ is of the order of 1 mbar (101 Pa) at 60-70°C. Additionally, the authors argue that a fraction of H$_2$ produced by the anaerobic corrosion of Cu is absorbed by copper. According to these studies, the anaerobic corrosion rate of copper is about 5µm/year. (Hultquist*, 1986) (Hultquist, o.a., 2015)

These claims have, however, been countered in several studies and some studies trying to reproduce the results obtained by Hultquist have not been successful. Several explanations for the observed hydrogen production have been offered by other authors such as hydrogen originating corrosion of stainless steel components of the test cell and the outgassing of H absorbed into the copper during the manufacturing process. (Lilja & King, 2011). None of these explanations have thoroughly been satisfactory though as the same results have been acquired in the studies with different experimental setups. The debate on the possibility of copper corrosion in the absence of oxygen and sulfide is ongoing.
2.4 Radiation induced corrosion

The conditions in the of the deep granitic groundwater are reducing and the elements are in their reduced forms, such as iron is found to be in Fe$^{3+}$ instead of Fe$^{2+}$ and manganese in Mn$^{2+}$ instead of Mn$^{3+}$. However, there have been studies suggesting the possibility of formation of oxidizing agents, such as oxygen peroxide and OH radicals, due to radiolysis by gamma radiation passing through the copper canister. Even though the Cu layer is 5 cm thick, this is not enough to stop the gamma radiation. The free OH radicals and the hydrogen peroxide produced by radiolysis of water have higher reduction potentials than that of Cu, leading to a possibility of Cu being oxidized by the oxidizing agents resulting from radiolysis of water (Jonsson, 2012). These issues will be discussed further in chapter two of this study.

Apart from corrosion resistance there are other properties that are needed to ensure safety over a long period against various kind of possible hazards, such as withstanding isostatic and shear loads. The copper used in the canister is processed in a number of different ways at the canister laboratory in Oskarshamnn to enhance its mechanical properties such as creep ductility by addition of phosphorus. The canister is analyzed by NDT methods such as ultrasonic and x-ray testing to avoid any material defects within the copper structure. These issues have discussed in detail in technical reports by the Swedish Nuclear Fuel and Waste Management Company (Technical report TR-10-46, 2010)
3 Bentonite Clay

Bentonite clay is one of the engineered barriers within the KBS-3 concept for the final disposal of the Swedish high level nuclear spent fuel. Bentonite buffer is placed around the copper canister holding the spent nuclear fuel to protect it from the surrounding hydrogeological environment (figure 1). The main function of bentonite buffer is to limit the transport of dissolved corroding agents to the canister and, through swelling pressure, reduce the microbial activity at the surface of the canister as well limit radionuclide transport to the surrounding geosphere in case of canister failure and release of radionuclides. Properties of bentonite clay such as mechanical stability, pH buffering capacity, swelling in contact with water makes it a suitable material for protection of the canister in the KBS-3 concept (Salas, Sena, & Acros, 2014) (Karnland, 2010) (Acros, Bruno, & Karnland, 2003). In the following lines, the microstructure of the bentonite clay, its barrier functionality and processes that are relevant for its long-term stability and functionality are generally discussed.

![Figure 1: Schematic illustration of the deposition chamber bentonite buffer and the copper canister. Source: SKB](image)

3.1 Bentonite microstructure and composition

The geological term bentonite describes soil materials containing high amounts of swelling mineral, usually montmorillonite, which belongs to the smectite mineral group. One of the characteristics of minerals belonging to this group is the layered structure and swelling properties. Other common properties of the members of this group is that individual layers have thickness of about 1 nm and the other two directions may extend to hundreds of nanometers. Each layer is composed of three sheets; a central octahedral sheet linked to an upper and lower sheet through shared oxygen atoms. This is referred to as 2:1 layered structure. (Murray, 2006) (Karnland, 2010)
Montmorillonite is 2:1 phyllosilicate mineral with an alumina octahedral sheet sandwiched between two tetrahedral sheets of silica (figure 9). The octahedral sheet has aluminum as the central atom while the lower and upper tetrahedral sheets have silicon as the central atom. The aluminum in the octahedral site is partly substituted by (mainly) magnesium and silicon atoms in the tetrahedral sheets are replaced by aluminum. Thus, a negative charge (with charge density 0.13 Cm$^{-2}$) prevails the layers. This negative charge is balanced by adsorption of cations, such as Na$^+$ and Ca$^{2+}$, between the individual layers, the interlayer space. These elements in the interlayer space are exchangeable and this property, referred to as cation exchange capacity (CEC), is measured in terms of milliequivalents per 100 grams. Cation ion exchange capacity of smectite minerals is an important property in many of the clay mineral’s applications. (Muray, 2006) (Murray, 2006) (Karnland, 2010) (Salas, Sena, & Acros, 2014) (Glaus, Baeyens, Bradbury, & Andreas, 2007)

The interlayer bonding is by van der Waals forces and by cations. In case of water absorption, interlayer swelling takes place as the distance between the layers increase by uptake of water molecules leading to an increase in the total volume of the clay. The charge balancing ions are bound by electrostatic forces and maybe exchanged by other ions in hydrated state. (Karnland, 2010) (Murray, 2006)

The charge balancing cations have a great influence on a number of physico-chemical properties of bentonite. Therefore, the dominating cation is included in the name of montmorillonite to indicate the type, such as Na-montmorillonite. Both Ca-montmorillonite and Na-montmorillonite are the commonly occurring smectite minerals. However, Ca-montmorillonite is the most common type of montmorillonite and is found in many areas of the world. (Muray, 2006) (Karnland, 2010)

Karnland (Karnland, 2010) describes the individual layers in montmorillonite with the following formula:

$$Si_{8-x}Al_x Al_{4-y}Mg_y(Fe) O_{20}(OH)_4 c^v_{(x+y)/v}n(H_2O)$$

Tetrahedral layer octahedral layer interlayer cations

where: $x<y$ and $0.4<x+y<1.2$ and $v$ representing the mean valence of the charge compensating cations (c). (Karnland, 2010)
In commercial bentonites montmorillonite content is usually above 60 wt%. For use as buffer material, a 75-90 percent montmorillonite content, is desirable. The mineralogy of the remaining part of bentonite may vary depending on geochemical conditions under which the bentonite is formed. However, some clay minerals that are typically expected in bentonite may be feldspars, gypsum, quartz, calcite, pyrite and different oxides and hydroxides. The composition and distribution of the accessory minerals in bentonite have great influence over the long-term geochemical evolution of the buffer in the repository environment. (Karnland, 2010). In the KBS-3 repository, Wyoming bentonite (commercially called MX-80), will be used as the bentonite buffer. MX-80 consists of 75 wt% montmorillonite. Other minerals i.e. quartz (15%), feldspars (7%), carbonates (1.4%), sulphides (0.3%) and organic carbon (0.4%) make up the rest of bentonite clay mineral. (Rosberg & Werme, 2008)

Apart from montmorillonite and accessory minerals, bentonite may contain amorphous and organic compounds within its structure. Organic compounds are considered a rather difficult issue to handle since it is hard to predict their long-term behavior in the buffer. One of the important issues in terms of chemical composition of bentonite is identification of minerals that may have high solubility and tendency to diffuse out of the bentonite buffer. In the long-term perspective, dissolution and diffusion of these highly soluble species may decrease the buffer density and effect its swelling property. Sulfate minerals, such as anhydrite and gypsum, are such highly soluble minerals whose solubility is even higher in higher temperatures. Apart from its role in sulfates, sulfur is also important from the perspective of corrosion of the copper canister since sulfides corrode copper. (Karnland, 2010) (Salas, Sena, & Acros, 2014)

3.2 Ion exchange an important parameter

As the performance of the bentonite barrier is crucial for the overall function of the KBS-3 concept, the long-term evolution of bentonite clay in repository conditions have been investigated as part of the safety assessments over the years. Experiments have been carried out to investigate mineral transformations and alterations in porewater chemistry in bentonite as well the exchange and mobility of species in the bentonite microstructure. (Salas, Sena, & Acros, 2014) (Wallis, Idiart, Dohrmann, & Post, 2016).
The chemical and physical properties that make bentonite a suitable material as a barrier in the KBS-3 concept, may be altered by its interaction with the groundwater, which may lead to weakening of its capacity as barrier material. One of the main effects of contact with groundwater observed by (Wanner H., 1986) and (Wanner, Wersin, & Sieroo, 1992) in laboratory experiments is the substitution of Na by Ca in the bentonite and equilibration with accessory minerals, such as calcite, a mineral that controls the pH of the system. In addition, sulphate content was found to rise due to dissolution of gypsum and/or anhydrite in the bentonite. (Wanner, Wersin, & Sieroo, 1992) (Wanner H., 1986)

A modeling study by Acros et. al. (Acros, Bruno, & Karnland, 2003) confirmed that cation exchange will be the main geochemical process taking place in the bentonite in contact with groundwater. The modeling study was based on the bore-hole experiments, referred to as Long-Term Buffer Material (LOT), carried out in the Äspö hard Rock Laboratory (HRL). The purpose of LOT is to develop an understanding of the long-term behavior of the buffer material under repository-like conditions. The experiments are carried out in 7 boreholes which contain a heater in a copper tube surrounded by bentonite (MX-80) cylindrical blocks. Different measures are taken to simulate the repository conditions such as some of the bentonite blocks containing KCl and NaCl to simulate increased salinity in the groundwater while the heater simulating thermal power from the SNF. In addition, calcite and gypsum are added to some blocks to simulate the possible effects of common bentonite accessory minerals.

According to the modelling study by Acros et. al., the replacement of Na by Ca takes place because of gypsum dissolution in the groundwater and diffusion of the resulting Ca from the granitic groundwater into the bentonite. Apart from gypsum dissolution, Ca is released due to calcite dissolution, which increases the pH of the system. This is beneficial from the perspective of copper canister corrosion. However, experiments in which NaCl and KCl were included in the bentonite additives/granitic/groundwater system, a decrease of pH was observed. The release of Na and K because of dissolution of NaCl and KCl resulted in the exchange of Ca by these ions. The resulting high concentration of Ca in the solution led to precipitation of both gypsum and calcite, which in turn resulted in significant decrease of pH reversing the beneficial effect of calcite i.e. the increase in pH. (Acros, Bruno, & Karnland, 2003) (Wanner, Wersin, & Sieroo, 1992). These results indicate that even though calcite controls one of the most important parameters i.e. pH, other processes may lead to supply of excess Ca reversing the good effects of pH in terms of increase of pH. (Acros, Bruno, & Karnland, 2003)

3.3 Ion mobility and transport mechanisms

One of the main functions of bentonite buffer is to prevent the transport of contaminants and corrosive species towards the copper canister and retention of radionuclides in case of canister failure. It is thus important to study the mobility of different species in the bentonite barrier. (Rosberg & Werme, 2008)

The transport of the solutes in bentonite has been found to proceed through advection in the initial (re-saturation) phase i.e. after the placement of the SNF and backfilling with bentonite. After saturation, though, the main transport mechanism has been observed to take place through
molecular diffusion of the solutes. Diffusion as the transport mechanism after saturation has been explained as the result of the low conductivity of bentonite due to high degree of compaction (Wallis, Idiart, Dohrmann, & Post, 2016) (Salas, Sena, & Acros, 2014) (Tertre, Delville, Pret, Hubert, & Ferrage, 2015). Diffusion is the processes of flux of the chemical species (molecules and particles) in a solution due to presence of a chemical potential. According to Flick’s first law, the flux of a chemical specie $J_x$ in bulk water in x direction is proportional to its concentration gradient $\frac{dc}{dx}$:

$$J_x = D_w \frac{dc}{dx}$$

$D_w \text{ (m}^2\text{/s})$ is a proportionality constant referred to as the diffusion coefficient:

$$D_w = \frac{RT}{N_Af}$$

Where:

- $R$: gas constant
- $T$: the absolute temperature
- $N_A$: the Avogadro constant
- $f$: a friction constant

### 3.3.1 Pore diffusion model

The diffusion behavior of charged species in compacted bentonites is different from that in free water due due geometrical factors associated with the complex structure of the clay minerals. The physicochemical states of water present in smectite clays have been found to be different in nature depending on the location of the porewater. The properties of water found in the pore space between the sheets are similar to those of the bulk water while the those of water present in the interlayer region is effected by large amount of charge balancing cations. The bentonite clay could be considered a double-porous medium consisting conceptually of macropores through which “free water” diffusion takes place and nanopores formed by the interlayer water. This has been referred to as pore diffusion model or macropore/nanopore diffusion model. (Glaus, Baeyens, Bradbury, & Andreas, 2007) (Tertre, Delville, Pret, Hubert, & Ferrage, 2015) (Bourg, Spositor, & Bourg, 2008)

In order to assess diffusion properties of different species, studies are carried out using tracer elements in various experimental setup, such as through-flow experiments, in which a compacted swelling clay sample is placed between two aqueous reservoirs. In through-flow experimental setups, one of the reservoirs (the upstream) contains tracers of interest. After the flow of the upstream reservoir through the swelling clay, the diffusion coefficient is acquired by measuring the tracer species in the downstream reservoir and modelling the flux of the species through the mineral clay. Both simple and more sophisticated pore diffusion modes have been used to model the diffusion data for a range of different species. (Tertre, Delville, Pret, Hubert, & Ferrage, 2015) (Glaus, Baeyens, Bradbury, & Andreas, 2007) (Bourg*, Spositor, & Bourg, 2006)
In a study by Glaus et. al. the diffusion of $^{22}$Na and $^{85}$Sr were studied in highly compacted montmorillonite and kaolinite as a function of the concentration of salt (NaClO$_4$) in an external solution in equilibrium with the clay mineral. In the case of montmorillonite, it was found that the external salt concentration variation clearly influenced the transport of $^{22}$Na, both in terms of diffusion and retention properties. It was observed that diffusion coefficients calculated based on tracer concentration gradient in the external aqueous solution in equilibrium with the clay, and the sorption distribution ratios decrease with increasing salt concentration in the external solution. (Glaus, Baeyens, Bradbury, & Andreas, 2007)

Due to high density of negative charge, the diffusion of anions may be limited; thus, the diffusion of neutral species and cations is of much higher importance in both macropores and nanopores. (Glaus, Baeyens, Bradbury, & Andreas, 2007) (Tertre, Delville, Pret, Hubert, & Ferrage, 2015). However, this has been challenged in a relatively recent study by Hedström and Karnland (Hedström & Karnland, 2012) in which ion exclusion in interlayer space due to electrostatic repulsion forces is referred to as a “misconception”. Calculations based on Donnan equlibrium shows, according to the study by Hedström and Karnland, that when a compacted and confined montmorillonite is in thermodynamic equilibrium with an external NaCl solution, the concentration of NaCl in montmorillonite (interlayer space) decreases as compared to that in the external salt solution, but not totally excluded. Thus, there is still excess salt, i.e. both anions and cations, in the interlayer space. According to the study, both Donnan equilibrium and Poisson-Boltzmann equation could be used to calculated the amount of the excess salt in the interlayer space. Based on MD simulations, the authors argue that the interlayer is the main diffusion pathway for both anions and cations. (Hedström & Karnland, 2012)

Birgersson and Karnland (Birgersson & Karnland, 2009) showed through equations based on ion equilibrium between interlayer and external electrolyte solution that diffusion through interlayer space is the main transport path for the sodium and chloride ions and that there is no need for invoking any other clay porosity concept. The effective diffusion coefficient, according to this study, may be proportional to an ion equilibrium coefficient. In case of anions the equilibrium coefficient was calculated by applying basic Donnan equilibrium approach. (Birgersson & Karnland, 2009) (Hedström & Karnland, 2012)

3.3.2 Surface diffusion model
Another phenomenon referred to as surface diffusion has often been proposed as the mass transport mechanism for the diffusion of species in mineral clays. Surface diffusion in clay minerals maybe described as migration of species within the electrical double layer of the mineral surface. The concentration gradient of positively charged cationic species perpendicular to the mineral particle surface is increased due to their interaction with the negatively charged silicate minerals. The dissipation of the gradient is prevented by the electrostatic potential of the mineral surface. The flux of the species along the mineral surface may increase with increasing gradient. (Oscarson, 1994) (van Shaik, Kemper, & Olsen, 1965)

For the surface migration to occur, a significant sorption of the species to the surface is required. Surface diffusion cannot be expected to be the transport mechanism for the species tightly sorbed to the clay surface. It is therefore important to understand the equilibrium relationship between the sorbed species and those in the solution to understand surface migration of these species. The
bonding energies holding monovalent cations to exchange sites in the interlayer region are of the same order of magnitude as those holding cations in the network of water molecules and as thermal energy. This and the fact that the concentration of sorbed cations on the surface may be much higher than those in the bulk solution indicate that these (sorbed) ions may play a significant role in the transport process. (Shainberg & Kemper, 1965) (Oscarson, 1994)

It has been claimed in some recent studies that surface diffusion can be used to model published diffusion data covering a wide range of cations, clays and chemical conditions. (Gimmi & Kosakowski, 2011)

There have been more advanced models based on the pore and surface diffusion models such as those based on multi-porosity structure of the clay minerals (Ochs, Lothenbach, Wanner, Sato, & Yui, 2001) (Ochs, Talerico, Sellin, & Hedin, 2005) and those with other approaches such as Donnan and ion exchange equilibria (Birgersson & Karnland, 2009) (Hedström & Karnland, 2012) and inverse modelling (Samper, o.a., 2006). In addition, there have been numerous studies covering effects of different factors on the diffusion of species through swelling clay minerals. In these studies factors such as the influence of density (Wu, o.a., 2012) (Joseph, o.a., 2017) and pH/temperature (González Sanches, o.a., 2008) (Wu T., o.a., 2016) as well as effects of organic matter (Wu T., Wang, Zheng, & Jin Ying, 2015) and salinity (Lida, Yamaguchi, & Tanaka, 2011) (Villar, 2006) have been investigated for sorption and/or diffusion of a wide range of species such as Ca$^{2+}$ and various radionuclides, for example, U, P, Cs, I, Pd, Tc, Re and Se through swelling clay minerals have been studied. Covering all those issues is out of the scope and the focus of the study. The effects of radiation from SNF on the different properties of bentonite, will be addressed in chapter 2.

3.4 Colloids

Bentonite clay is the outermost of the three engineered barriers in KBS-3 concept. While in contact with the copper canister on the inner side of the barrier, the outer side is in direct contact with the bedrock. This means that the porewater of bentonite will mix with the groundwater in the surrounding rock formation. It has been observed that bentonite’s contact with water coming from the rock formation leads to clay hydration, swelling and formation of a surface gel. Under appropriate conditions, the surface gel can transform into a sol. Therefore, corrosion of different origins (chemical and physical) may occur at the gel front leading to release of particles in the range of 1nm - 1μm, referred to as colloids. These colloids, moving freely in the water, may be potential contaminant carriers in groundwater (Albarran, o.a., 2014) (Alonso, Albarran, García-Gutiérrez, & Cormenzana, 2011). The erosion of the bentonite, which has been reported to occur at the bentonite buffer/granitic rock interface, depends on the physico-chemical conditions, such as gas and water pressure, bentonite compaction, groundwater flow velocity, pH and ionic strength of the groundwater. In the scenario of glacial water intrusion, the high pH and low salinity of the water, properties that favor colloid/particle release from bentonite, may lead to accelerated erosion of the bentonite buffer. (Bouby, o.a., 2010) (Alonso, Albarran, García-Gutiérrez, & Cormenzana, 2011)

The chemistry of the groundwater influences to great extent the stability of colloidal particles. The stability of montmorillonite colloids has been found to decrease with increasing ionic strength and
decreasing pH. The effects of temperature are also influenced by pH and ionic strength. At lower pH values (pH<4), it has been found that increasing temperature reduces the stability of montmorillonite suspensions while at higher pH values (pH≥10) an increase in temperature leads to stabilization of the colloidal suspensions. At intermediate pH values, the effect of temperature proved to be dependent on the ionic strength while at lower and higher temperature values the effects of temperature were independent of the ionic strength. (García-García, Jonsson, & Susanna, 2006)

3.4.1 Transport of colloids in fractured rocks

Na-montmorillonite is more prone to colloid formation as compared to Ca-montmorillonite as the swelling capacity of Ca-montmorillonite, the property that induces gel propagation, is lower than that of Na-montmorillonite. The colloids released from the bentonite surface may enter open rock fissures, joints or available pore space. The transport behavior of colloidal particles in fractured rocks may be fundamentally different from that in the porous media such as clay minerals and soils due to the difference in the geometry of the transport of conduits. Fracture transport of colloidal particles may be more significant owing to higher physical shear forces and larger conduits under intense infiltration (Zhang, Xiangyu, Weisbrod, & Guan, 2012) (Weisbrod, Ofer, & Eilon, 2002). The transport of colloidal particles in rock fractures is mainly influenced by advection, adsorption/desorption, dispersion, physical straining and air-water interface capturing (figure 3). (Zhang, Xiangyu, Weisbrod, & Guan, 2012) (Weisbrod, Ofer, & Eilon, 2002)

![Figure 13: Processes involved in colloid transport in fractured rocks (Zhang, Xiangyu, Weisbrod, & Guan, 2012)](image)

Even though colloid transport could be rapid in the fractured rocks, there has been indications it may retarded by some factors, in some cases significantly. The main retention mechanisms preventing the transport of the colloids in natural fractures and fissures are the colloid sedimentation, attachment to surfaces and filtration (figure 4). The capability of colloidal particles to aggregate and become larger in size influence their deposition rate since larger and heavier
particles settle under gravity. Filtration processes depend on the size of colloidal particle; the transport in fractured rocks of smaller particles have been shown to be influenced by Brownian diffusion while that of larger particles may be affected by settling. (Albarran, o.a., 2014)

![Diagram of colloidal retention mechanisms in fractured rocks](Garcia-Garcia, Jonsson, & Susanna, 2006)

Figure 14: Colloid retention mechanisms in fractured rocks (Garcia-Garcia, Jonsson, & Susanna, 2006)

The mobility of the colloids in groundwater is controlled by particles’ aggregation and sorption capabilities. Radionuclides with low solubility in groundwater maybe be particularly prone to colloid-borne migration since these radionuclides would otherwise be much less mobile due to their high sorption capability. DLVO theory, named after Derjaguin, Landau and Overbeek, can be applied to make predictions about the mobility of inorganic particles/colloids. According to this theory, the total interaction energy between to colloids/particles is the sum of an electrostatic repulsion energy, $V_R$, arising from the overlap of electrical double layers and an attractive energy, $V_A$, due to van der Waals-London dispersion forces:

$$V_T = V_A + V_R$$

However, considerable discrepancies have been observed between predictions based on DLVO theory and experimentally measured values when conditions are unfavorable for coagulation and sorption, for example when particles and surfaces have the same charge. DLVO theory was originally developed for smooth bodies with ideally geometries and uniform surfaces which is the reason for its short coming in covering non-ideal conditions such as rough and non-uniform surfaces with heterogenous compositions. The DLVO theory has therefore been subject to modifications and developments in order to include other factors and forces involved in the dynamics of particle interaction. (Swanto, 1995) (Alonso, Missana, Patelli, & Rigato, 2007).
Chapter Two
Possible radiation effects on the engineered barriers

1 Introduction to the chapter
In this chapter, a radiation-focused analysis of the KBS-3 concept is carried out to determine the regions within and in the respective surroundings of the engineered barriers where radiation from the SNF may initiate processes relevant for the integrity of the barriers. In order to be able to make assessments regarding the effects of the enhanced radiation in the identified regions, an understanding of the type of radiation occurring at these spots and their respective variation in intensity during the different periods of time over the life-span (10^5 years) of the KBS-3 repository is important.

Chapter two is divided into two sections; in section 1, an attempt is made to identify the regions within the KBS-3 design where enhanced radiation may occur. In section 2 of the chapter, an analysis of the possible/potential effects of radiation in the identified regions is carried out. This analysis is based on the current knowledge of the radiation-induced corrosion and radiation-induced microstructural damages of materials of the engineered barriers and contents of the copper canister.

In chapter three, an attempt is made to evaluate whether all the possible radiation-induced damages discussed in this chapter have been considered in the overall safety analysis of the KBS-3 repository.

2 Regions under enhanced radiation
The two main engineered barriers i.e. the copper canister (and its contents) and the bentonite barrier will be within the range of the radiation field from the SNF. In the following subsections, a closer look is taken at the regions where enhanced radiation may occur and lead to radiation-induced processes of relevance for the safety analyses of the engineered barriers.

2.1 The fuel
A description of the characteristics of SNF has been given in chapter one. In order to identify the regions within the fuel assemblies where enhanced level of radiation may occur and cause radiation-induced processes, an understanding of the design and dimensions of the fuel assemblies is required. In addition, the material composition of the fuel assembly needs to be considered while assessing the effects of radiation on the structural materials (metals and metal alloys) of the fuel assemblies.

An illustration of the BWR and PWR fuel assembly design provided to SKB by nuclear fuel suppliers is given in figure 1. The BWR fuel assemblies contain between 60 to 100 fuel rods which consist of zirconium alloy tubes filled with cylindrical UO_2 fuel pellets. The length of a BWR fuel assembly is about 4.3 m and its cross-sectional area is about 0.141×0.141 m^2. The fuel
rods are arranged in square arrays emplaced in a fuel channel. With a cross-sectional area of 0.214x0.214 m$^2$ and the total length of about 4.3 m and the number of fuel rods ranging between 204 to 264, PWR fuel differ from a BWR fuel in terms of design and dimensions. This has consequences for the design of the canister and the insert (see below under the subsection “the canister”). (Technical report TR-10-13, 2010)

Figure 2: Fuel assembly design and dimensions of BWR (left) and PWR (right). (Technical report TR-10-13, 2010)

There have been studies, although limited in number, on the effects of radiation on the UO$_2$ fuel under repository conditions. However, the studies covering long terms effect, those relevant for deep geological repository time scales, have been limited in number. One of the most important aspects of the effects of the radiation emanating from the SNF, is the variation in intensity of different radiation types over different periods of time in the repository.

As mentioned above, the material composition of the fuel assemblies is needed to assess what kind of materials are irradiated by SNF and effect of the radiation on the materials. As can been seen in table 2, the structural materials used in the PWR and BWR fuel assemblies mainly consist of zirconium alloys, stainless steel and nickel alloys. All these materials will be within the radiation field of the SNF.
2.1.1 Materials within the radiation field
Radiation originates from the SNF and occurs within the structure of the fuel as well as at the free rod volume i.e. the plenum volume and the pellet-cladding gap. The metal alloys used as structural materials in the fuel assemblies, such as zirconium alloys and stainless steel in the cladding, fuel channels and other parts of the assembly such as bottom and top plats, spacers etc., will be exposed to the radiation from the SNF. The radiation doses these materials will receive depends upon the burn-up of the fuel and the age of the SNF (table 2).

<table>
<thead>
<tr>
<th>Fuel age (years)</th>
<th>Dose rate (Gy s⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>38 MWd kg⁻¹</td>
</tr>
<tr>
<td>10²</td>
<td>1.19 × 10⁻¹</td>
</tr>
<tr>
<td>10³</td>
<td>2.87 × 10⁻²</td>
</tr>
<tr>
<td>10⁴</td>
<td>5.90 × 10⁻³</td>
</tr>
<tr>
<td>10⁵</td>
<td>5.44 × 10⁻⁴</td>
</tr>
</tbody>
</table>

Table 4: Alpha doses from nuclear fuel with two different burns ups (Nielsen & Jonsson, 2006)

The geometrical dose distribution i.e. radiation dose rate as a function of distance from the fuel surface are the most relevant aspects that need to be considered. As the yield of radiolysis
products differ for different types of ionizing radiation (Jonsson, 2012), it is important to distinguish between the dose rates of the different radiation types i.e. alpha, gamma and beta. Thus, the type and the range of the radiation in the air and, in the case of water-intrusion into the canister, in the water are some aspects that need to be taken into account.

Water intrusion into the canister within the first 1000 years of the deposition is not considered a realistic approach (Jonsson, 2012). This means that in the relevant safety analyses, a repository age of at least 1000 years should be considered. In this case, alpha radiation will be the dominating type of radiation within the immediate vicinity of the SNF. However, as the range of alpha radiation is short in both air and water (maximum range in water 40 μm and a few centimeters in air), most of the groundwater in contact with the fuel, in case of water intrusion, will be subjected to gamma and beta radiation doses. However, alpha radiation will cause formation of radiolysis products in the immediate proximity of the fuel. The formation and effects of radiolysis products is discussed in section 2.

2.2 The insert
The insert is made of a class of cast iron referred to as nodular iron which is expected to provide the whole canister the required mechanical strength and stability against different kinds of loads. Examples of such loads may be those emerging from pressure of water in the depths of 500 meters and the pressure exerted by the bentonite clay when it swells. One of the characteristics of cast iron is its higher carbon content (usually over 2%), which is present as graphite in the structure of the cast iron. Graphite may occur in different forms in cast iron affecting its material properties significantly. The kind of cast iron used for the insert (nodular iron) has graphite in spherical forms, which gives it good mechanical properties. The form, size and distribution of the graphite in cast iron can be modified by the addition of magnesium and other elements. The nodular cast iron for the insert will have an Fe content of more than 90%. Additionally, alloying elements such as nickel, manganese, chromium and copper may be added to enhance its material properties. (SKB, 2013) (Technical report TR-10-46, 2010)

As for the design of the insert, it contains individual channels for each fuel element. The BWR insert is designed with twelve channels while there will only be four channels in the insert for PWR fuel assemblies (figure 3). Both designs will be about 949 mm in diameter leading to a total outer canister diameter of 1050 mm (figure 2). In order to allow space for the canister shell to be able to deform plastically inwards in case of any mechanical force or load exerted on its surface and due to fabrication-related reasons, a clearance of a few millimeters between the shell and the insert is maintained. (SKB, 2013) (Technical report TR-10-46, 2010)

From the perspective of radiation shielding, it may be interesting to note that the thickness of the cast iron between the fuel-insert and insert-copper interfaces is not homogenous; the minimum-thickness points, the distance is 38.3 and 43.2 mm for BWR and PWR assemblies, respectively
Figure 3: Cross section of BWR and PWR inserts and its dimensions (Technical report TR-10-13, 2010)

The canister will be filled (90%) with argon changing the atmosphere from air to argon. In addition, the fuel assemblies in the insert channels may contain some water despite the drying processes before the encapsulation fuel elements. According to SKB, the water content, in the most pessimistic assumption, may be as high as 600 g per canister. (Technical report TR-10-46, 2010) (SKB, 2013)

2.2.1 Materials within the radiation range
The materials in the insert, such as iron and copper and other alloying elements mentioned above, will be within the field of the radiation from the SNF. In addition, the argon gas replacing the air in the canister and water content in fuel assemblies will also be receiving radiation doses.

2.3 The copper shell
A five-centimeter-thick shell of high-purity, oxygen-free copper surrounds the insert in the canister design for the KBS-3 repository. The choice of copper as the outer shell, according to SKB, is due to its high resistance to corrosion with properties satisfying the specifications in /EN 1976:1988/ for the grades Cu-OFE (UNS10100). The chemical composition of the Cu-OFE is given in table 3; the additional requirements are the following: 30–100 ppm phosphorous, < 0.6 ppm hydrogen, < 12 ppm Sulphur. Oxygen in very low concentrations (some tens of ppm) may also be allowed.
Table 5: Chemical composition of Cu-OFE (European Copper, 2012)

<table>
<thead>
<tr>
<th>Chemical composition</th>
<th>Value</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag [ wt.% ]</td>
<td>0.0025</td>
<td></td>
</tr>
<tr>
<td>As [ wt.% ]</td>
<td>0.0005</td>
<td></td>
</tr>
<tr>
<td>Bi [ wt.% ]</td>
<td>0.0002</td>
<td></td>
</tr>
<tr>
<td>Cd [ wt.% ]</td>
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<td></td>
</tr>
<tr>
<td>Cu [ wt.% ]</td>
<td>99.99</td>
<td></td>
</tr>
<tr>
<td>Fe [ wt.% ]</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>Mn [ wt.% ]</td>
<td>0.0005</td>
<td></td>
</tr>
<tr>
<td>Ni [ wt.% ]</td>
<td>0.001</td>
<td></td>
</tr>
<tr>
<td>P [ wt.% ]</td>
<td>0.0003</td>
<td></td>
</tr>
<tr>
<td>Pb [ wt.% ]</td>
<td>0.0005</td>
<td></td>
</tr>
<tr>
<td>S [ wt.% ]</td>
<td>0.0015</td>
<td></td>
</tr>
<tr>
<td>Sb [ wt.% ]</td>
<td>0.0004</td>
<td></td>
</tr>
<tr>
<td>Se [ wt.% ]</td>
<td>0.0002</td>
<td></td>
</tr>
<tr>
<td>Sn [ wt.% ]</td>
<td>0.0002</td>
<td></td>
</tr>
<tr>
<td>Te [ wt.% ]</td>
<td>0.0002</td>
<td></td>
</tr>
<tr>
<td>Zn [ wt.% ]</td>
<td>0.0001</td>
<td></td>
</tr>
</tbody>
</table>

The canister is basically a copper tube sealed with a lid and a bottom using a welding technology referred to as friction stir welding (FSW). In this technology, a rotating tool is used to join the canister tube to the lid and the bottom. The rotating tool stirs the copper material and causes high temperatures (850 °C) heating around the joint merging the parts together (figure 3). (SKB, 2013)

Figure 4: The principle of friction stir welding (SKB, 2013)
2.3.1 Materials within the radiation range
The copper shell (its microstructure) and its welded parts will be within the reach of the radiation from the SNF. In addition, alloying metals in Cu-OFE will also be receiving radiation doses. According to SKB, the dose rate at the surface of the copper canister will not be in excess of 1 Gy/h (ca $3 \times 10^{-4}$ Gy s$^{-1}$). This will be ensured through the choice of fuel assemblies from the interim storage facility and measuring of radiation levels before encapsulation. This, according to an estimation, means that the dose rate will be about three orders of magnitude lower than the dose-rate at the surface of the SNF after 100 years. As gamma radiation mostly originates from the Cs-137, which has a half-life of 30 years, the dose rate at the surface of the copper shell will be decreasing by 50% every thirty years. (Jonsson, 2012)

Another ferrous material in contact with the copper and the insert will be the insert steel lid. Apart from Fe, the steel lid contains small amounts of C, Si, Mn, P and S (Andersson, Eriksson, & Westman, 2004).

2.4 Radiation within the bentonite barrier
The structure and chemical composition of bentonite has been described in chapter two. The montmorillonite particles may also contain iron as a substituting atom for aluminum in the octahedral sheet as well as in other forms.

Apart from building-block materials of montmorillonite and the exchangeable cations, the water in the interlayer region and pore water in the compacted bentonite will be under irradiation from SNF. The dose rate at the copper-bentonite is expected to be 0.1-0.5 Gy/h at the canister deposition time (Håkansson, 2000) (Holmboe*, Mats, & Susanna, 2012).

3.5 Radiation at the interfaces
Apart from the regions mentioned above, there are a number of interfaces where the engineered barriers are in contact with each other. The interface between the fuel assembly (the cladding) and the insert, the space between the insert and the copper shell, and the interface between copper and the bentonite will all be within the field of radiation. The contact of the dissimilar metals in terms of reduction potentials may be relevant from the perspective of corrosion as redox conditions may be changed by the water radiolysis products (refer to sections below).

3 Possible effects of SNF radiation
In this section, the possible and/or potential effects of the radiation from SNF on the materials within the regions identified in section 1 will be discussed. As for copper canister, there are basically two possible scenarios which this study is based on: In scenario 1, the copper canister is intact and maintains its barrier function i.e. the corrosion of copper is not severe enough to lead to penetration of water into the canister and the radionuclides do not escape into the biosphere. In scenario 2, the canister is damaged and water enters the canister and comes in contact with the fuel elements. It is to be noted that scenario 1 will be the case until and if scenario 2 becomes a reality i.e. the occurrence of scenario 1 does not necessarily mean that scenario 2 will not take place. The effects of radiation on the engineered barriers and their respective surroundings are discussed with respect to both scenarios. In the end of this chapter, all the possible radiation
induced damages to the engineered barriers discussed in this chapter are summarized and categorized under scenario 1 and scenario 2.

3.1 The fuel
The long-term evolution of the spent fuel in the repository has briefly been discussed in chapter one. Since the fuel is already subjected to very high radiation doses and dose rates during the reactor operation, it may be argued that the effects of radiation on the fuel itself and the construction materials used in the fuel assembly may be of little significance. However, the time-scales to be considered in safety assessments for geological repositories are different from those relevant for the safety analysis of reactor operations. In addition, granitic groundwater has a chemical composition which is different from the coolant/moderator used in nuclear reactors. The composition of the groundwater may further be changed during the passage through the bentonite structure and incorporation of species within the bentonite structure. In scenario 2, this may be an important factor to be considered in the analysis of radiation effects on the fuel.

3.1.1 Radiation induced-dissolution
In scenario 2, the water will absorb radiation from the SNF which will lead to formation of radicals and ionic species from water molecules. As mentioned above, both the fuel itself and its construction materials will be within the reach of the radiation from the SNF. The locally produced radicals and ionic species have the capability to alter the redox conditions fundamentally. In pure water, the radiation from the SNF leads to the formation of the both oxidants and reductants: HO•, H•, HO2, solvated electron e\textsubscript{aq}−, H+, H2, and H2O2. If the water contains other solutes, such as in the case of granitic groundwater, the radicals formed due to radiolysis of water will react with the solutes to form other radicals. This may prevent the radical-radical reactions, a process which leads to termination of initially formed radicals. Only solutes present with higher concentration will be able to prevent radical-radical reactions leading to a change in their G-values. However, the reaction of radicals with the solutes may lead to formation of other reactive species such as the formation of carbon-centered radicals under oxygen free conditions in case of dissolved organic substances. These reactions may alter the water chemistry of the groundwater affecting the dissolution of fuel.

Over the recent years, a number of studies have been performed to understand the mechanisms involved in oxidative dissolution of the spent fuel by water radiolysis products. The identification of H2O2 as the main oxidant and its catalytic decomposition on the surface of the UO2 have been important observations for the long-term safety analysis of the repository. The role of fission products in oxidative dissolution and the studies involving UO2 doped with different species to mimic the spent fuel have offered new insights into mechanisms involved in the radiation-induced dissolution of SNF. However, studies addressing radiation-induced dissolution, especially those addressing processes at the solid-liquid interfaces have been very limited in numbers. For example, the G-value of H2 at metal oxide-water interface has been shown to be much higher than those in the bulk water. No such discrepancy has, however, been observed for H2O2 (Jonsson, 2012). More studies are needed to investigate these discrepancies and other related issues.

3.1.2 Radiation-induced structural changes
In case of contact with the ground water, the spent nuclear fuel (mainly UO2) will undergo complex alterations processes. Under oxidizing conditions, the nearly insoluble U(IV) is oxidized
to the much more soluble U(VI)-complexes (Sureda, o.a., 2011) (Hanson, o.a., 2005). As the concentration of these U(VI)-complexes increase, it may result in precipitation of secondary phases of U(VI).

The near-field radiation from the SNF may have a profound effect on the fuel itself and its alteration products. According to a number of studies, the secondary phases of U(VI) may be prone to different kind of structural damages as a result of exposure to, for example, electron radiation. As beta particles in general are not energetic enough (0.1 – 1.5 MeV) to cause displaced atoms by ballistic processes, electron radiation has been used in some studies to simulate β-decay events. The crystalline structure of most of these complexes were found to collapse (amorphization) with radiation doses in the range of $10^{10}$ Gy. (Sureda, o.a., 2011)

One of the parameters effecting the amorphization dose of the alteration products of UO$_2$ has been shown to be the variation in temperature. Due to radioactive decay of SNF, the temperature in the near-field of the repository will initially be as high as 250 °C decreasing to below 100 °C within a few hundred years. The amorphization dose of one of such alteration products, uranophane (Ca(UO2)2(SiO3OH)235H2O, and soddyite ((UO$_2$)$_2$(SiO$_4$)(H$_2$O)$_2$), increased from $10^{10}$ Gy to $10^{11}$Gy under temperatures above 140 °C. This dependence of radiation stability on the variation in temperature was interpreted to be associated with the elimination of structural water from the alteration products. (Sureda, o.a., 2011)

The amorphization of these alterations of UO$_2$ may have a significance for the nuclide-retention properties of the spent fuel as these U(VI)-complexes such as uranophane and soddyite ((UO$_2$)$_2$(SiO$_4$)(H$_2$O)$_2$) have the capacity to retain radionuclides such as cesium and strontium as well as, technetium, neptunium and selenium (Douglas, Clark, Utsunomiya, & Ewing, 2002) (Chen, Burns, & Ewing, 1999) (Chen*, Burns, & Ewing, 2000) (Buck, McNamara, Douglas, & Hansonq, 2003) (Friese, Douglas, & Jerden, 2006) (Burns, Deely, & Skanthakumar, 2004).

The intrusion of water into the canister within the first 1000 years of the deposition is considered to be an unrealistic scenario (Jonsson, 2012) which may mean that there is a limited possibility of formation of secondary phases of UO$_2$. The possibility of canister breach after 1000 years may still lead to formation of secondary phases of UO$_2$. In this regard, the alpha radiation close to the surface of the fuel may cause radiolysis of water layer and change to the redox conditions. It is thus necessary to study the radiation-induced amorphization of the UO$_2$.

3.1.3 Induced radioactivity in crud
Crud is layers of deposited corrosion products on different spots in the reactor system. Corrosion of metals in different parts of the reactor vessel lead to dissolution of species such as iron and zinc in the circulating coolant of reactor. These corrosion products deposit on different parts of the reactor system including the nuclear fuel pin surfaces leading to a number of problems including heat flux issues and accelerated oxidation (corrosion) of BWR and PWR fuel cladding (Water Chemistry Control in LWRs, 2012) (Corrosion of Zirconium Alloys, 2012). Some of the corrosion products become radioactive due to neutron radiation from the nuclear fuel during the reactor operation, which can lead to exposure of maintenance workers to high radiation doses during shutdown. $^{60}$Co is the major source of out-of-core radiation for maintenance
workers. The source of $^{60}$Co is $^{59}$Co which is found as impurity in the nickel in structural alloys, such as 304 stainless steel, used in the reactor system. Apart from $^{60}$Co, which is responsible for 80-90% of out-of-core radiation, species such as $^{54}$Mn, $^{58}$Co, $^{59}$Fe and $^{65}$Zn also contribute to radioactivity (Corrosion of Zirconium Alloys, 2012) (Water Chemistry Control in LWRs, 2012). The extent of neutron activation depends on neutron flux intensity, neutron absorption cross-sections and the duration of exposure to neutron flux (Water Chemistry Control in LWRs, 2012). As neutron and other types of radiation will not seize after the deposition of the canisters, the extent of effects of radiation from these sources should be considered.

3.2 The insert
The insert can be considered one of the barriers within the canister with a very important function to keep the fuel by providing the canister the required mechanical strength. As in case of other metal and metal alloys in the canister components and the fuel assemblies, the insert will be exposed to radiation affecting its microstructure and, in the case of water intrusion, to the radiolytically produced radicals and other reactive species.

3.2.1 Radiation induced microstructural damage
It is a well-known fact that radiation with sufficient energy affects the microstructure of metals leading to significant microstructural alterations. Atoms are arranged in three dimensional periodic arrays referred to as crystal lattice in metals. An illustration of the three possible structures i.e. simple cubic (sc), face centered cubic (fcc) and body centered cubic (bcc) is given in figure 5.

![Three possible crystal structures](image)

*Figure 5: Three possible crystal structures: the simple cubic (sc), body-centered cubic (bcc) and centered cubic (fcc) crystal structures.*

The letter $a$ on each side of the structures denotes the length of a side of the unit cell, known as a lattice parameter.

Radiation induced microstructural alterations starts with a particle, for example a neutron with a certain energy or a high-energy photon, colliding with an atoms nucleus in the radiated metal. On receiving enough radiation energy from the colliding particle, the atom is displaced from its crystal lattice site resulting in a vacancy that’s left behind by the atom. The recoiling atom, if it possesses enough, energy may displace another atom in the lattice site by collision leading to formation of another vacancy. Depending on the energy of the initially received energy, this process may continue leading to a collision cascade. Every displaced atom, after dissipating its energy, will eventually come to rest within the crystal lattice as a self-interstitial defect. The number of displaced atoms and the those of the vacancies left behind will essentially be the same forming Frenkel pairs (figure 5). The number of Frenkel pairs formed is also referred to as the
number of displacements. The accumulated density of the Frankel pairs is expressed as the number of displacements per atom (dpa). If this number reaches one, it means that each atom has been displaced once on average. (Wolfer, 2012) (Zinkle, 2012)

![Figure 6: Displaced atom forming a Frenkel pair (Lattice defects and diffusion, 2017)](image)

The vacancies and self-interstitials that are formed due to displacements of atoms may diffuse through the crystal during the high temperatures under reactor operations. Thus, the vacancies and self-interstitials will encounter each other leading to either annihilation or formation of vacancy and interstitial clusters. In addition, they may also be captured in other extended defects, for example cavities, dislocations and grain boundaries.

Since the number of dislocations and vacancies are equal (Frenkel pairs), one may argue the numbers of interstitials and vacancies that arrive at each sink would eventually become equal. Thus, one may expect that the microstructure of sinks should eventually reach a steady state leading to no further microstructural changes due to continuous radiation. However, the contrary has been found to be the case; the radiation-induced void swelling has been found to continue without reaching steady state affecting the macroscopic properties of the metals (Wolfer, 2012).

The effects of radiation from the SNF on the microstructure of the metals in the insert is thus essential to be considered in the safety analysis of the canister in the KBS-3 repository concept. As far as the metals used as construction materials in the fuel assembly, such as stainless steel and zirconium, they have already been subjected to high levels of irradiation during the reactor operation. The insert, however, will be exposed to the irradiation once the fuel assemblies are placed in the canister. Having gone through a different manufacturing process (cast iron), its properties are different from those of the metals in the construction material of the fuel assembly. For example, the copper content of the cast iron of insert may be prone to precipitation and cluster formation due to radiation effects making the cast iron brittle and less ductile which may cause it to break. Thus, any studies addressing the effects of radiation on the fuel assembly materials during the reactor operation may not be suitable for use to assess the possible or potential radiation-induced, long-term damage to the insert. Besides, the insert will be exposed to a different dose rate (lower) under a very long time as compared to the other materials in the reactor during the operation.
3.2.2 Radiation induced Cu-precipitation
The precipitation of Cu particles in $\alpha$-Fe, as mentioned above, is a familiar phenomenon which has been studied extensively. However, the studies addressing this issue have been concerned with the phenomenon occurring during the life-time of the reactor, a time-scale which is much shorter in comparison to time-scales relevant for geological repositories. The radiation-induced precipitation, which depend on the Cu content in the iron, may lead to embrittlement of the metal. As there is a certain amount (<0.05 at.%) of Cu in cast iron used for the insert and the fact that the bulk of the material (cast iron) will be within the radiation-field from the SNF, it’s important that the phenomenon be studied from the perspective of long-term radiation under the repository conditions. The dose rate the cast iron will be exposed to during the repository storage will be considerably lower as compared to the dose rate metals are exposed to during the reactor operation. However, as the temperature in the repository environment will also be lower as compared to the reactor temperature, there will be a difference in the formation and annihilation of the defects in the cast-iron insert material (Brissonneau, Barbu, & Bocquet, 2004). Taking into accounts these issues, there is a need to consider this issue in the safety assessments of the copper canisters.

3.2.3 Radiation induced corrosion
As far as scenario 2 (water-intrusion into the canister) is concerned, radiation may have effects on the insert material (cast iron) in terms of corrosion as the water chemistry is altered significantly by the radiolytically produced species and their reactions with the solutes in the groundwater. These effects should, however, be analyzed in a broader perspective i.e. in the light of other processes taking place in the case of water intrusion into the canister, for example, the effects on the anaerobic corrosion of the ferrous materials (cast iron and steel) as radiation has been shown to affect the rate of anaerobic corrosion of steel. (Smart, Rance, & Werme, 2008)

It is to be noted that before and/or if scenario two takes place, the insert and other materials in the canister will have been receiving high doses of radiation for hundreds or thousands of years. Even if one assumes that the radiation effects are not significant enough to cause a failure during scenario 1, one may evaluate the possibility of corrosion, for example SCC, when scenario 2 becomes a reality. Any safety analysis should therefore take into consideration the fact that in case of water-intrusion, the corrosion processes will be not be affecting a newly manufactured cast iron insert, rather one that has already by exposed to radiation for a very long time. Apart from the initial high doses of radiation the insert will be receiving, it has been shown that ferrous materials may be prone to microstructural damages due to long-time low dose radiation (Cole & Allen, 1998).

3.2.4 Water and argon in the canister
Before the they are placed into the canister, the fuel assemblies are dried by applying well-established drying processes to ensure low enough water content in the canister. However, some water does remain in the fuel assemblies after encapsulation leading to the question whether radicals and other species produced due to radiolysis of water could be an issue to consider in the overall safety assessments. According to SKB, the water content will not exceed 600 g in the canisters. Thus, the amount of residual water may not be enough to be significant in terms of
radiation induced corrosion. However, a very recent study of copper corrosion revealed that corrosion in humid argon atmosphere may be significantly higher than that in anaerobic water (Björkacka, Johnson, Leygraf, & Jonsson, 2017). It is thus important to consider this possibility in the safety assessment of the engineered barriers. Further investigations of radiation-induced corrosion of copper and ferrous materials (field such as cast iron and stainless steel) in argon atmosphere are thus essential to understand these, for the KBS-3 safety analyses, very relevant processes.

3.3 The copper shell
The outer surface of the copper shell will be in contact with bentonite clay and its contact with groundwater will be independent of the two scenarios this study is based on. The extent of contact with water of the outer surface of the copper shell will depend on the possibility of breach of bentonite clay’s barrier function. The kind of events can lead to such a breach have briefly been discussed in chapter one. The possibility of radiation induced corrosion of the copper shell will thus be discussed from both perspective of availability of groundwater at the surface (and consequently the availability of radiolysis products) as well as the microstructural damage due to radiation from the SNF.

3.3.1 Radiation induced corrosion
Two of the radiolytically produced oxidants, i.e. H$_2$O$_2$ and HO·, have much higher reduction potentials (E°(·OH$_{aq}$/H$_2$O) = 2.59 V vs. SHE, E°(H$_2$O$_2$/2 H$_2$O) = 1.77 V vs. SHE) than copper (E°(Cu$^+$/Cu(s)) = 0.520 V vs. SHE, E°(Cu$^{2+}$/Cu(s)) = 0.341 V vs. SHE) (Björkacka, Johnson, Leygraf, & Jonsson, 2017). The radiation induced corrosion of copper under anoxic conditions is thus thermodynamically feasible.

Even though the dose rate at the surface of the copper shell will be considerably lower than the dose rate at the SNF surface, water will still be decomposed by the radiation from the SNF leading to formation of the water-radiolysis products mentioned above. While considering radiation induced processes that may have a relevance for the integrity of the copper shell, a holistic approach is needed. In this regard, the conditions prevailing, such as groundwater chemistry, redox conditions, temperature, pH and salinity and dose rate variations, in the repository environment should be considered. The effects of temperature on the radiation induced processes and those of the dose rate have, to a very limited extent, been studied. A few studies have also been carried out to investigate the role of different solutes in the groundwater in terms of radiation effects. It has for example been shown that the chemistry of water influences the reactions that the radiolytically produced species may undergo as well as the dose rate and the total dose may influence the corrosion of the copper shell. In a study by Björkbacka et. al., it was found that gamma radiation may cause significant corrosion of the copper shell under anoxic conditions. (Jonsson, 2012) (Björkbacka, Hosseinpour, Leygraf, & Jonsson, 2012)

Earlier studies performed to investigate radiation induced corrosion of copper has been carried out under different dose rates and total doses leading to different results. According to an estimation by Håkansson (Håkansson, 2000), the outer surface of the copper shell will have received a maximum total dose of about 100 kGy within the first hundred years after the
deposition. In a relatively recent study (Björkbacka, Hosseinpour, Johnsson, Christopher, & Jonsson, 2013), the effect of total dose received by copper samples from SKB under anoxic conditions was investigated. It was found that after receiving total doses of 35.5 and 74 kGy respectively, the oxide layer thickness of the copper samples was 50-100 nm while the thickness of oxide layer of non-irradiated (reference) samples was about 4 nm. In addition, a significant difference was observed in terms of copper concentrations between the irradiated and non-irradiated samples in the solutions when the samples had received a total dose of 74 kGy. A correlation between the dissolution of copper during irradiation and the total dose received by the copper samples was observed.

These studies may, however, be insufficient to understand the complex nature of radiation induced processes that are very relevant to the safety analysis of the copper canister. Studies addressing radiation induced corrosion of copper have been few in numbers. Several issues relevant for the radiation induced processes that may influence the integrity of the canister need more investigations. (Jonsson, 2012)

3.3.2 Radiation-induced microstructural defects
The copper properties can be changed by the damage due to gamma and neutron radiation during the lifetime (10^5 years) of the repository. Although a non-ferrous metal, the microstructure of copper is prone to these changes as gamma radiation can interact with crystalline structures in different ways. Apart from gamma rays, the SNF emits neutrons by spontaneous fission and \( \alpha \)-particles emitted from recoiling nuclei. An \( \alpha \)-particle formed during a decay may be absorbed by an oxygen atom in a UO\( _2 \) molecule. In this process, the oxygen isotope is transmuted into neon leading to emission of a neutron. The heavier nuclei generated by these processes may interact with the structure of the copper shell through ballistic processes.

In a short-term perspective, the microstructural changes due to gamma radiation may be insignificant. However, as the copper shell will be exposed to radiation from the SNF during the life-time of the repository (10^5 years), it is important to consider such changes in safety analysis of the KBS-3 repository. The first step in such an investigation may be quantifying the source and the emission yields from the SNF or accumulative doses the copper shell will be exposed to during the life-time of the repository. By applying the available scientific knowledge in the field, such as displacement theory, and well-established scientific processes, it is possible to then evaluate the radiation-induced damage rate for the copper shell over the repository life-time.

3.3.3 Radiation effects on FSW joints
FSW is a well-studied technology for both same-metal joints and hybrid joints (joints between two different metals), mostly for applications in the automobile industry (Rao, Naik, & Srinivas, 2017). Effects of different factors and parameters have been studied on the weld morphology and microstructure (Besharati-Givi & Asadi, 2014) (Pierpaolo & Palazzo, 2014) (Miličić, Gladović, Bojanić, Savković, & Stojić, 2016) (Yaduwanshi, Bag, & Pal, 2017). Depending on a number of factors, such as the tool rotational and travel speed, variation in axial force, applied load and the position of the probe tip, the resulting weld may differ from each other in terms of mechanical properties (Sun & Fujii, 2010) (Miličić, Gladović, Bojanić, Savković, & Stojić, 2016).
Despite the knowledge acquired from the research and applications in the industry, the response of copper FSW joints to long-term radiation is poorly studied. The copper shell is probably the most important of the engineered barriers in KBS-3 design; FSW joint need to have corrosion-resistance properties as close as possible to the parent metal. It is thus essential that the effects of long-term radiation on the microstructure of the joints and effects of water radiolysis products on the corrosion of the joints be studied in detail.

3.4 The bentonite clay
As far as the effects of radiation or radiation induced processes on the bentonite barrier is concerned, under scenario 1 no radionuclides will be released from the canister and the possibility of the transport of radionuclides through the barrier is irrelevant. However, as scenario 1 does not exclude the possibility of the occurrence of scenario 2, it is of interest to make an assessment about the effects of direct radiation and/or radiation induced processes on the microstructure of bentonite before a breach occurs and scenario 2 is the case. This can be seen as asking oneself whether bentonite’s exposure to radiation during a long period of time will have altered its properties to an extent that it may affect the transport of the radionuclides through the bentonite if/when scenario two becomes a reality.

3.4.1 Radiation induced effects
As mentioned in chapter one, Montmorillonite is 2:1 phyllosilicate mineral with an alumina octahedral sheet sandwiched between two tetrahedral sheets of silica (figure 9 in chapter one). The octahedral sheet has aluminum as the central atom while the lower and upper tetrahedral sheets have silicon as the central atom. The aluminum in the octahedral site is partly substituted by (mainly) magnesium and silicon atoms in the tetrahedral sheets are replaced by aluminum. The montmorillonite particles may also contain Fe, mostly as a substituting atom for aluminum in the octahedral sheet, but also in other forms. It may be found as iron oxides sorbed onto the surface of the layer and as hydroxyl complexes on the layer surface located the edge of the montmorillonite layer. In the KBS-3 repository, Wyoming bentonite (commercially called MX-80), which has a 3 wt% Fe content, will be used as the bentonite buffer.

The composition and distribution of the accessory minerals in bentonite have great influence over the long-term geochemical evolution of the buffer in the repository environment. The oxidation state of the different species may have influence on different properties of montmorillonite. The water-radiolysis products formed by radiation from the SNF may alter the redox conditions in water in the bentonite and at the copper-bentonite interface leading to changes in oxidation states of the different species. It was for example found in a relatively recent study that ɣ-radiation with doses within the levels expected at the bentonite surface may alter Fe(II)/Fe_{tot} ratio. The oxidation state of iron in the crystal structure of smectite minerals changes their physico-chemical properties. The alteration of Fe(II)/Fe_{tot} ratio by radiation from the SNF may greatly influence a number of physical properties, such as cation exchange capacity (CEC), swelling pressure, total layer charge and specific surface area, of montmorillonite.

One of main oxidants among the water-radiolysis products is H_{2}O_{2} which undergoes different processes at a solid-liquid surface in relation to the processes taking place in the bulk water. It has for example been reported that H_{2}O_{2} adsorb on the surface of metal oxides such as UO_{2} and
undergoes catalytic decomposition. It has also been noted that H$_2$O$_2$ may be associated with the changes in the observed Fe(II)/Fe$_{tot}$ ratio under $\gamma$-radiation. The role of radiolytically produced H$_2$O$_2$ in the bentonite pore and layer water content as well as at the copper-bentonite interface needs to be investigated further. (Stucki, Lee, Zhang, & Larsson, 2002) (Jonsson, 2012) (Holmboe*, Mats, & Susanna, 2012)

3.4.2 Radiation-induced amorphization

One of the issues relevant for the safety analysis of the bentonite buffer is the radiation induced crystalline to amorphous transition of clay minerals. Over the recent decades this phenomenon has been investigated in several studies. This is an issue important for the radionuclide retention properties of smectite clays as cumulative doses may alter the properties such as sorption, swelling or water retention of clay minerals. It is thus essential for the safety analysis of the bentonite barrier that its response to radiation be investigated properly.

The dose rate and energy of the particles the results of the studies so far have been based on differ from those the bentonite will be exposed to in the KBS-3 repository. In some studies, for example, alpha radiation has been used to study the response of clay minerals which may not be relevant for the KBS-3 repository under scenario 1 as the range of alpha radiation may not be long enough to reach the bentonite barrier. However, in scenario 2, the possibility of radionuclides escaping the canister cannot be excluded which necessitates the investigation of effects of radiation (including ballistic processes) from the escaped radionuclides on their respective surroundings including the bentonite clay.

In some other studies, conditions expected in the repository have not been considered properly. For example, bentonite response to electron radiation is studied under much higher temperatures than those expected at the surface of the bentonite clay in the KBS-3 repository.

As amorphization has a significant influence over the dissolution kinetics of smectite, and thus on its radionuclide-retention properties, it is important that the behavior of the montmorillonite clay under exposure to long-term radiation be considered in safety analyses. (Sorieul, o.a., 2008) (Fourdin, o.a., 2010) (Meunier, Velde, & Griffault, 1998) (Haire & Beall, 1979) (Wang, Wang, W.L., & Ewing, 1998)

3.5 Processes at the interfaces

There are several interfaces in the KBS-3 design where different materials, within the radiation field, come in contact with each other. The insert will be in contact on the inner side with the fuel cladding (cladding-insert interface) and on the outer side with the copper shell (insert-copper interface). The bentonite clay will be in contact with copper (copper-bentonite interface) on the
inner side while its outer surface will be contact with the bedrock (bentonite-copper interface). The possible radiation effects at these interfaces are discussed briefly in the follow subsections.

3.5.1 Metal-metal interfaces
Corrosion is an electrochemical processes and redox conditions in a corrosion system play an essential role on the possibility and extent of corrosion. In scenario 2, the first two interfaces i.e. cladding-insert and insert-copper (figure 6) as well as steel lid’s contact with copper shell where metals and metal alloys with different standard potentials are in contact with each other under the radiation field from the SNF may be important from the perspective of corrosion. How the water radiolysis products effect the corrosion processes is thus very important to study in detail. Studies addressing these aspects should cover a range of different conditions, redox potentials, pH, salinity, concentrations of different solutes in the granitic groundwater etc., which may be expected during different periods during the life-time of the KBS repository. In addition, it is important that the effect of radiation on different properties of bentonite, such as compaction capability and radionuclide retention, be investigated in detail as these properties are important for different functions the bentonite barrier is expected to offer.

Figure 7: Illustration of cladding-insert and insert-copper interfaces (Source: Originally SKB with additions by the author)

3.5.2 Copper-bentonite and bentonite-rock interface
As far as the copper-bentonite and bentonite-bedrock interface (figure 7) are concerned, there may be possibility of processes (other than corrosion) being influenced by the irradiation from the SNF. A number of issues associated with possible processes at the copper-bentonite interface can be relevant for the safety analysis of the KBS-3. In scenario 2, the radiolytically produced species may interact with both the bentonite and the copper. Irradiation of water in the pore and interlayer region close to the bentonite’s inner surface will lead to formation of water radiolysis products close to the copper-bentonite interface. The radiation-induced processes taking place at this
interface may be effected by the chemistry of the bentonite-equilibrated groundwater. These processes may be relevant for the corrosion of copper and the properties of the bentonite clay. It is thus important that detailed studies of the such processes are conducted to understand and categorize the possible relevance of these processes to the integrity of the copper shell and the bentonite clay.

As for the bentonite-rock interface, contact with groundwater from water-bearing fractures in the bedrock may cause the bentonite to disperse into particles referred to as colloids. A number of factors in the groundwater such as temperature, pH and salinity influence the rate of formation of such colloidal particles. If the conditions are favorable for the processes to continue, it may cause the volume of the bentonite to decrease as the bentonite is transported away continuously from its surface in contact with the groundwater. In scenario 2, the released nuclides may adsorb on the surface of these colloids and be transported into the geosphere.

![Diagram of copper-bentonite and bentonite-bedrock interfaces](Source: SKB illustration with additions by the author)

The effects of the factors mentioned above on formation of colloids have been studied in some detail. By applying the DLVO theory, as mentioned in chapter one, the effects of pH, temperature and ionic strength on the stability of colloids can be qualitatively predicted. However, effect of ionizing radiation on the stability of the colloidal particles have poorly been studied.

The stability of the colloids dispersion can be affected by $\gamma$-irradiation; it has for example been found that $\gamma$-irradiated Na$^+$-montmorillonite colloids exhibit higher stability as compared to the unirradiated colloidal particles. The stability of the colloids can be caused by change in the surface potential of the colloidal particles due to interaction with water radiolysis products. It is also possible that $\gamma$-irradiation interacts directly with the bentonite colloidal particles (Holmboe, Wold, Jonsson, & Garcia-Garcia, 2009). These observations are highly relevant for the safety analysis of
the bentonite barrier. It is therefore important that more detailed studies be carried out to understand the phenomena.

3.6 Bacterial activity
A third possible scenario would be the failure of the bentonite barrier function. A brief account of the phenomenon leading to such a scenario is given in chapter one. Such a failure may cause less or no compaction pressure and the availability of anoxic groundwater at the bentonite-copper interface. There has been evidence of diverse and active populations of microorganism in the granitic rock aquifers down to depths of 1500 m (Pedersen, 1999). It is thus theoretically possible that microorganism may exist within the bentonite structure even if no breach of the bentonite compaction function occurs. However, in the case of the breach, the availability of water at the copper surface may allow bacterial activity. Since radiation may have certain effects on sulphate reducing bacteria (SRB), which are associated with anoxic corrosion of copper (Bruhn, o.a., 1999), it is important that radiation effects on such microorganism under repository conditions be investigated thoroughly.

4 Summary of the possible effects
There are mainly three types of irradiation induced damages relevant for the safety analysis of the engineered barriers i.e. the canister (the fuel, the insert and the copper shell) and the bentonite clay:

i. The radiation induced microstructural defects
ii. The radiation induced crystalline to amorphous transition
iii. Oxidation/corrosion/dissolution due to redox changes induced by radiolytically produced species

The first type of damage is relevant for metals such as the cast iron used as the insert material and stainless steel in the fuel assemblies as well as the insert steel lid. The second type is relevant for ceramic and clay minerals, mainly the UO$_2$-fuel and the bentonite clay used as compacting and backfill material. The third type of the radiation induced damage i.e. possible damages due to redox changes caused by radiolysis products formed by radiation from the SNF is relevant for both the canister (and the content within) and the bentonite clay.

Two scenarios, i.e. scenario 1 where the canister maintains its barrier function and scenario 2 in which water enters the canister, have been the basis for this study. The possible processes leading to radiation-induced damages are summarized under the respective scenario in the following lines:

4.1.1 Scenario 1: In the case of the canister maintaining its barrier function
- The long-term effects of radiation and radiation induced processes on the bentonite clay
- Oxidation/corrosion of Cu by radiolytically produced oxidants outside the canister
- Radiation-induced microstructural damage of the copper shell and separate investigation of FSW joints in terms of radiation-induced damage.
- Radiation-induced microstructural damage of the bentonite clay
- Radiation-induced microstructural damage of the (cast-iron) insert
• Radiation-induced (long-term) microstructural damage of the fuel cladding material

4.1.2 Scenario 2: In the case of failure of the canister barrier function

In case of water-intrusion into the canister, the effects of radiation on the engineered barriers must be seen in the light of other processes that maybe ongoing. The effects of radiation on the microstructure of the metals in the canister (different ferrous materials and the copper) during scenario 1 should be considered in assessing the effects of radiation in/during scenario 2.

• Effects of radiation on the structure of secondary phases (alterations of U(VI)-complexes) which may have formed due possible oxidizing conditions caused by radiolytically produced oxidants
• Oxidation/dissolution of UO\textsubscript{2} matrix due to processes initiated by the radiolytically produced oxidants
• Radiation-induced corrosion of the fuel cladding
• Radiation-induced corrosion of the (cast-iron) insert
• Effects of radiation on the bentonite clay
• The effect of radiation on the anaerobic corrosion of ferrous materials

Apart from the two scenarios discussed above, a third scenario would be the breach of bentonite barrier function. In this scenario, the effects of radiation on the sulphate reducing bacteria (SRB) at the surface of copper will also be important to study.
Chapter three
KBS-3 radiation issues – SKB’s work so far

1 Introduction to the chapter
The purpose of this chapter is to discuss whether the radiation issues summarized in chapter two have been considered in the overall safety analysis carried out by SKB so far and/or whether they are included in the future research and development plans for KBS-3 system. The assessment is mainly based on the information in the relevant documents made available by Swedish Nuclear Fuel and Waste Management Co (SKB) and information in open literature such as scientific studies by the academia.

The discussions and assessment in this chapter are based on the radiation issues mentioned and discussed in the SR-site main report, the underlying SKB process and technical reports and/or future research and research programs mentioned in FUD-programs. An introduction to these reports and why they are considered relevant for this study is given in the following section.

2 Identifying the relevant documents
In 2011, SKB submitted a safety assessment report, referred to as SR-Site main report, as part of the application to Land and Environment Court to build the repository for the final disposal of Swedish spent nuclear fuel in Forsmark. The purpose of the SR-Site is explained in the following words in the first of the three volumes of the report:

The SR-Site report is a main component in SKB’s licence application to construct and operate a final repository for spent nuclear fuel at Forsmark in the municipality of Östhammar. Its role in the application is to demonstrate long-term safety for a repository at Forsmark. (SR-Site, 2011).

In addition, the purpose of the SR-Site has been explained in several SKB reports to be the safety assessment of KBS-3 repository and to provide feedback in terms of, inter alia, future needs of research for SKB’s research and development program. According to, for example, the Data report for the safety assessment SR-Site (TR-10-52), the following are the main purposes of the safety assessment project SR-Site:

- To assess the safety, as defined in applicable Swedish regulations, of the proposed KBS-3V repository at Forsmark;
- To provide feedback to design development, to SKB’s R&D programme, to detailed site investigations and to future safety assessment projects. (Technical report TR-10-52, 2010)

Moreover, the SR-Site report presents the current knowledge and the status of the development of the relevant technologies for the KBS-3 system. In addition, an evaluation of the remaining uncertainties in relation to protection of human beings and the environment against the hazards of radiation after the deposition of the canister is made. (SR-Site, 2011).
The SR-Site 2011 and the SKB process reports for SR-site and other technical reports are the basis of the discussion in this study. The technical reports are produced by SKB experts and are based on scientific studies, such as those conducted by academia or within the framework of other nuclear waste management programs, for example the geological repository in the Yucca Mountain Nuclear Waste Repository (King, Ahonen, Taxén, Vuorinen, & Werme, 2002) (SR-Site, 2011). Some technical reports have been the result of SKB’s contracts with expertise outside the company, such as the TR-01-32 (Radiation effects in spent nuclear fuel canisters) developed by Michael W Guinan (Guinan, 2001).

An illustration of the hierarchy of SKB’s documents i.e. the SR-Site main report, and additional references to the SR-Site is given in figure 1. For the description of the different documents mentioned in figure 1, the reader of this report is referred to Data report for the safety assessment SR-Site (TR-10-52).

Two of the three process reports mentioned in figure 1, The Fuel and canister process report and the Buffer, backfill and closure process report are the main documents used to evaluate SKB’s work relevant for this study till 2011. These two reports are referred to as TR-10-46 and TR-10-47, respectively and contain discussions concerning the processes, including radiation-induced processes, relevant for the safety assessment of the KBS-3 repository design. According to the SR-Site main report, these process reports, developed in support of the SR-Site 2011, are based on other SKB technical reports and open literature (scientific articles etc.) as well as on SKB’s FEPs (Features, Events and processes – refer to SKB’s Data report for details):

*The identification and handling of processes of importance for the long-term evolution and safety of the repository is a key element in the safety assessment. The identification of 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 three dedicated Process reports, one for the fuel and canister, one for the buffer, backfill and repository closure and one for the geosphere.

For the purpose of simplification, an illustration of the hierarchy of the documents relevant to this study is given in figure 2.

![Diagram](image)

**Figure 10:** Illustration of hierarchy of the SKB-documents relevant for this study. Description of the FUD-programs and other documents mentioned in figure 2 are given under the relevant sections in this report.

As far as the fuel and the canister are concerned, the SKB Fuel and canister process report (Technical report TR-10-46, 2010) is the basis for SR-Site as mentioned in the report:

_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-Site._

(Technical report TR-10-46, 2010)

As for the bentonite buffer, the SKB TR-10-47 (Buffer, backfill and closure process report for the safety assessment SR-Site) is the main report this study is based on (Technical report TR-10-47, 2010). In case of updates since SR-Site 2011, such as in the SKB TR-10-46 mentioned above, this work will be based on the updated versions of these reports.

There are basically two ways the processes mentioned and discussed in the SKB process reports (TR-10-46 and TR-10-47) are handled in SR-Site: either neglected on the basis of the information discussed in the report or included by means of modelling (Technical report TR-10-46, 2010) (Technical report TR-10-47, 2010). According to the SR-Site main report:

_The handling of all processes in a process report is summarised in a process table describing if the process is neglected, if it is quantitatively modelled or if the choice between neglect and modelling is subject to a specified condition that may or may not be fulfilled as the repository_
system evolves. Several of the processes are thus handled through quantitative modelling, where each model in general includes several interacting processes, often occurring in different system parts and hence described in different process reports. The models form a network, where results from one model are used as input to another. The network is described graphically by two Assessment Model Flowcharts, AMFs, and two associated AMF tables linking the processes in the process tables, the models in the AMFs and the reporting of the modelling exercise in this main report. (SR-Site)

As mentioned above, any processes identified and documented in the two Process reports mentioned above will be seen, in this study, as a process considered by SKB in the overall safety assessment procedure regardless of the handling in the SR-Site. A discussion regarding the thoroughness of the issues covered by these reports is carried out in the discussion section.

2.1.1 Documents related to future R&D
Since the SR-Site in 2011, there have been more scientific studies concerning the effects of radiation on the engineered barriers and different parts of the KBS-3 system after the closure of the repository (Björkbacka, Hosseinpour, Leygraf, & Jonsson, 2012). It is thus important to consider SKB’s plans for the research and development within the framework of the KBS-3 system. In this regard, the FUD-program 2016, in which SKB’s research and development efforts for the period 2017-2022 are presented is one of the FUD-programs for the discussion in this report. FUD-programs are developed and presented by SKB every three years; the latest in this series since the first FUD-report in 1986 is the FUD-program 2016. The FUD-programs preceding and following the SR-Site directly are FUD-program 2010 and FUD-program 2013, respectively. Apart from the system for the final deposition of the Swedish spent fuel, the FUD-programs address research and development needs for handling of short- and medium-lived nuclear waste as well as decommissioning of nuclear reactors and other nuclear plants. (Fud-program, 2016)

Apart from development of the required technologies and methodologies for the final deposition of the Swedish spent nuclear fuel, the purpose of the research and development program is to contribute to a better understanding (Sv. ökad processförståelse) of the processes relevant for the safety of the KBS-3 system (Fud-program, 2016). It is thus relevant for this study to make an analysis of whether all significant research outcomes related to the processes mentioned in chapter two of this study have been considered by SKB while planning the future research efforts. As the FUD-programs contain summaries of the current knowledge of the processes relevant for the long-term safety analyses of the KBS-3 repository design, these programs will be the basis of the analyses in this regard.

The focus in the FUD-program 2016, as far as the final deposition of Swedish nuclear spent fuel is concerned, is on the issues SKB has identified (and presented in SR-site) as significant for handling of the spent nuclear fuel. The future needs and plans for research and development within the framework of the final repository for the Swedish spent fuel have been addressed in section 5.4 and the associated subsections of the report. (Fud-program, 2016)
3 Radiation issues
In order to be able to make assessments concerning the effects of the radiation from the SNF on the engineered barriers, a thorough understanding of the nature of the radiation emanating from the SNF is needed. In the following section, a brief overview of SKB’s work in this regard is presented.

3.1 Understanding the characteristics of SNF-radiation
Studies covering the nature of the radiation from the SNF should encompass an understanding of the type of radiation occurring in different regions within the engineered barriers and their respective surroundings. This requires characterization of the radiation within the KBS-3 system as a function of time and space. In this regard, neutron and gamma fluxes calculated by Håkansson at Studsvik Nuclear AB (Håkansson, 2000) on behalf of SKB can be mentioned. This study has been the basis for some of SKB’s work on the effects of neutron and gamma radiation on the insert and the copper shell (Guinan, 2001). Both studies, however, were carried out back in 2000/2001 which may call for a renewed and updated understanding of the related issues. As for the updated information regarding the radionuclide inventory and the resulting radiation types, this requirement has been acknowledged by SKB in FUD-program 2016. In section 5.4.3. (Bränsleinformation, kriticitet och kärnämneskontroll), SKB points out the need for methods to determine any parameters of significance for the spent fuel such as fissile material, radionuclide inventory, burnout, decay time and reactivity etc. For example, the calculation code Scale which is one of the tools used by Håkansson in the study for evaluation of dose rates of neutron and gamma radiation will be updated and validated (Fud-program, 2016).

3.1.1 Radiation attenuation by the engineered barriers
The radiation attenuation by the engineered barriers has been addressed in the Fuel and Canister Process report as a process (process Radiation attenuation). The process is important from the perspective of evaluating of the dose rates at the different regions within and in the surroundings of the engineered barriers (refer to chapter two). The process is summarized both in Fuel and Canister and Buffer Process reports (TR-10-46 and TR-10-47, respectively). According to these reports, most of the neutron and gamma radiation will be attenuated by the cast iron and copper in the canister:

\textit{Gamma and neutron radiation from the canister are attenuated in the buffer...The maximum dose outside the canister has been calculated to be less than 500 mGy/h at time of encapsulation. The dose is dominated by Cs-137, which has a half life of ~ 30 years. The radiation that is not attenuated reaches as far as near-field rock. The main part of the gamma radiation from the spent fuel is shielded by the iron and copper in the canister. Only a minor fraction will reach the buffer.} (Technical report TR-10-47, 2010)

4 The main radiation phenomena
In chapter two of this study, the possible damage caused to the engineered barriers by radiation was analyzed in respect to two main types of radiation induced processes: indirect effects of radiation through the water radiolysis products and effects of direct radiation on the microstructure of the engineered barriers. In the lines below, SKB’s work in this regard is discussed to evaluate whether these phenomena have been considered in the overall procedure for the safety assessment.
4.1 Water radiolysis
The issue of formation of radical oxidants and reductants due to radiolysis of water has been addressed, for example, in SKB technical Report TR-10-46 (Fuel and canister process report for the safety assessment SR-Site). Based on a study by Allen (1961), the report mentions in section 2.5.3 (water radiolysis) the production of H₂ and H₂O₂ in the following words:

Radiolysis of water has been thoroughly studied by both experiments and modelling. Low LET (Linear Energy Transfer) radiation, such as gamma radiation, does not produce any detectable amounts of the stable species H₂ and H₂O₂ in pure de-aerated water /Allen 1961/, because they react via OH· and H· radicals in a chemical chain reaction to reform H₂O. (Technical report TR-10-46, 2010)

A number of studies have been referred to in the report, it is further stated that gamma radiolysis of water in the presence of small amounts of dissolved hydrogen may be very limited:

Recent experimental and radiolytic modelling studies on 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. (Technical report TR-10-46, 2010)

The effect of dose rates has been summarized in the following words:

Available information shows that there is no evidence for enhanced corrosion rates caused by gamma radiation. On the contrary, at least for dose rates in the range of 10–100 Gy/h, the experimental data seem to indicate a lower corrosion rate in the presence of radiation /King et al. 2010/. (Technical report TR-10-46, 2010)

As the issue of water radiolysis has been mentioned and discussed in a SKB process report, it has thus been considered in the SKB safety analysis according to criterion set in this study. As the formation of the water-radiolysis is considered by SKB, a summary of how SKB assesses the effects of water-radiolysis process on the engineered barriers is presented in the lines below.

4.1.1 The fuel
The possibility of radiation induced dissolution of the spent nuclear fuel in case of water intrusion has been mentioned and discussed under section 2.5.5 (Fuel dissolution) of the Fuel and Copper Process report. According to an estimation made in the report with reference to a study by Kolár and King (1996), any oxygen remaining in the groundwater after deposition and closure will be consumed by oxidation of copper approximately within a year. Any possibility of the oxidative dissolution depends, according to the report, on the redox conditions altered by radiolysis products. (Technical report TR-10-46, 2010)

The report states that water intrusion into the canister is not expected to be taking place within the first 1000 years after closure. Therefore, the main form of radiation will be the alpha decay. Due
to very short range in water, the alpha radiation will cause radiolysis in a very thin (about 35μm) layer of water in the vicinity of the fuel surface. However, in the presence of anaerobically corroding iron and its corrosion products, alpha radiation level after 1000 years is not expected to cause any significant oxidative dissolution of the UO₂. (Technical report TR-10-46, 2010)

A rather detailed account of the processes affecting the dissolution of UO₂ is given in the report with references to studies dealing with oxidative dissolution of the UO₂ based on SIMFUEL and UO₂ doped with different nuclides. For example, reference is made to studies by Jonsson et al. where the effect of H₂ on the fuel dissolution is discussed:

The results of spent fuel dissolution modelling, including the effect of Fe(II) ions on the decrease in steady state H₂O₂ concentrations at the fuel surface and the fuel surface reduction process on α-particles, have been published recently /Jonsson et al. 2007/. Calculation of maximum fuel dissolution rates using this model shows that an H₂ pressure of 0.1 bar is sufficient to effectively inhibit the dissolution of spent fuel aged 100 years or more. In the presence of 1 μM Fe(II), even 0.01 bar H₂ will be sufficient to stop fuel dissolution, while for Fe(II) concentrations of 40 μM (as expected inside a failed canister in Sweden), even radiolytically produced hydrogen is sufficient to completely inhibit fuel corrosion /Jonsson et al. 2007/. (Technical report TR-10-46, 2010)

According to the report, the process of fuel dissolution will be handled in the following way in the safety assessment (SR-Site):

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 UO₂ 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. (Technical report TR-10-46, 2010)

Radiation-induced oxidative dissolution of the spent fuel has thus been considered as a phenomenon by SKB.

4.1.2 Insert and copper
Indirect effects (via water radiolysis process) of radiation is mentioned both in Fuel and canister process report (TR-10-46) and Buffer, backfill and closure process report for the safety assessment SR-Site (TR-10-47). In the case of TR-10-47, the radiation-induced corrosion (by water radiolysis process) of the outer surface of the canister has been discussed. According to the report, the formation of the radiolysis oxidants and hydrogen, if not removed from the system, will cease as the equilibrium will soon be reached. Based on a study by Christensen and Pettersen (1997), an estimation of the extent of copper corrosion on the outer canister surface is made. According to this estimation, based on 5mm water layer surrounding the canister, the corrosion depth will be about 16 μm after 327 years. A reference has been made to experimental studies carried out by King et al. in 2001 which indicate that the overall effect of radiation on copper corrosion will be negligible under oxidizing conditions (Technical report TR-10-46, 2010) (Technical report TR-10-47, 2010).
The effects of the composition of porewater is acknowledged; however, the process is excluded from the SR-Site as the dose rate, according to the report, will be too low to lead to formation of enough oxidants:

*Dose rates are too low to cause any significant radiolysis outside canister – the process is excluded from SR-Site* (Technical report TR-10-47, 2010)

The effects of radiation on cast iron insert has been discussed under section 3.4.6 of the Fuel and Canister Process report. The discussion focuses on the possible radiation-induced microstructural defects of cast iron. As for the effects of radiolysis, no mention of this issue is found under section 3.4.6.

The phenomenon of indirect effects of radiation (via water radiolysis) on the canister has thus been considered as a process in the overall safety assessment procedure by SKB. The effects of radiation-induced corrosion of cast-iron has not specifically been discussed.

### 4.1.3 Radiation-induced corrosion in argon atmosphere

The canister will be filled with argon changing the atmosphere from air to argon in the canister. The effects of radiation on copper corrosion in water saturated argon atmosphere has been discussed in chapter two, section 3.2.4 of this study. Radiation induced corrosion of the ferrous materials (cast iron and steel) and copper under argon atmosphere has not been mentioned or discussed in the SKB process reports or SR-Site main report.

### 4.1.4 Bentonite buffer

The indirect effects (via water-radiolysis process) has been mentioned and discussed under section 3.5.13 of the TR-10-47 (Buffer, backfill and closure process report for the safety assessment SR-Site). The issue of formation of water radiolysis products by gamma radiation of the porewater has been discussed in respect to copper corrosion at the copper-bentonite interface. However, no mention of the effects of water radiolysis products and the consequent processes on montmorillonite has been made in the report (Technical report TR-10-47, 2010). Neglecting of the issue in the buffer process report is probably based on the calculation by Lundgren 2004 (SKB technical report TR-10-47) according to which the dose rate will be too low:

...[radiolysis of the porewater] could affect the canister surface, but also this effect is shown to be insignificant... The radiation field outside a canister has been calculated in /Lundgren 2004/. The calculated dose rate is lower than 500 mGy/h (100–150 mGy/h) at the time of deposition. (Technical report TR-10-47, 2010)

Indirect effects (via the water radiolysis process) on the bentonite buffer have thus not been mentioned and discussed by SKB, possibly owing to, according to the report, very low radiation doses due to radiation attenuation by the engineered barriers.
4.1.5 Effects on colloid stability
In chapter 2 of this study, the effect of radiation on the stability of colloids formed at the bentonite-bedrock interface has been discussed. In SKB’s Buffer and backfill process report, the issue of montmorillonite colloid release and the effects of different parameters on the phenomenon has been mentioned and discussed in section 3.5.11 of the report. The possible effect of radiation on the stability of these colloidal particles have, however, not been mentioned. The radiation intensity as a factor affecting the montmorillonite colloid release is considered in the report to be irrelevant (Technical report TR-10-47, 2010). The issue has thus not been considered by SKB in the overall safety assessment procedure.

4.2 Microstructural damages due to direct radiation
A number of issues related to the microstructural damages caused by direct radiation to the engineered barriers have been discussed in chapter 2. In this section, an account of SKB’s work in this regard is presented.

4.3 Effects on SNF
One of the effects of radiation on the SNF, the amorphization of alterations products of the fuel, leading to deteriorating radionuclide retention properties of these alterations, is discussed in chapter two (refer to section 3.1.2 of chapter two of this study). This phenomenon is not specifically mentioned by SKB in the Fuel and Canister Process report; however, the conditions i.e. the possibility of alteration of redox conditions close to the fuel in case of water intrusion into the canister, has been discussed. It is argued that neither in the case of alpha radiation close to the fuel surface nor the gamma radiation within the first 1000 years after closure, the radiolytically produced species may cause any significant changes to the redox conditions.

As for radionuclide release from crud in case of contact with water, the issue has been mentioned in Fuel and Canister Process report. However, the radiation effects arising from these released nuclides has not been discussed. As the dose-rates from the radionuclides released from the fuel is assessed by SKB to be too low to cause any significant radiolysis, there may not have been enough reason to consider the release of lower amounts of crud radionuclides an issue.

4.3.1 Effects on the insert cast iron
Guinan carried out a calculation of neutron and gamma displacement damage to the two main metals i.e. copper (fcc structure) and iron (bcc structure) used in the KBS-3 system under contract with SKB. The study is in part based on the neutron and gamma fluxes calculated by Håkansson at Studsvik Nuclear AB (Håkansson, Beräkning av nuklidinnehåll, resteffekt, aktivitet samt doshastighet för utbränt kärnbränsle (R-99-74), 2000). In addition, the results of the study were compared to earlier experimental studies covering this issue. There were a number of computer codes employed to determine the total defect production rates in the mentioned metals. The neutron damage code SPECTER from Argonne National Laboratory, EPDL (Evaluated Photon Data Library) and electron damage code DISP from Lawrence Livermore National Laboratory were used to determine the possible damage caused by neutron and gamma radiation, respectively.
According to the Guinan, the effect of short-term radiation on the metals would be insignificant. The main purpose of the study was thus to calculate the long-term effects (upto 100,000 years) of radiation on the microstructure of the metals:

*If we were dealing with ordinary time scales, we could immediately dismiss the possibility of any materials radiation effects. However, for the spent fuel canisters we must consider times up to 100,000 years. If the rates above persisted for the entire time, we could expect measurable, but not necessarily compromising, radiation effects in both iron and copper. The accumulated damage, however, is significantly reduced by both the further decay of the source of the radiation and thermal annealing as the damage is produced. (Guinan, 2001)*

However, according to the study, the long-term effects (gamma and neutron displacement damage) are insignificant:

*... we were able to conclude that the magnitude of any physical property changes, e.g. yield stress, creep rates, enhanced solute segregation, dimensional changes, or brittleness, resulting from exposure over the service life of the of the canister will be negligible. (Guinan, 2001)*

Another issue mentioned in chapter two in terms of microstructural damages, is the copper precipitation phenomenon (refer to section 3.2.2 of chapter two of this study). This has been addressed under section 3.4.6 (Radiation effects) of Fuel and Canister Process Report; reference has been made to the study by Brissonneau et al. (2004) and several others:

*Precipitation of copper particles is a well-known problem in reactor vessels (see e.g. /Odette and Lucas 1998, Chaouadi and Gérard 2005/). Depending on the Cu content in the material, precipitation may occur as a result of radiation, potentially leading to embrittlement of the material. Such an accelerated ageing process occurs gradually over the lifetime of the reactor; i.e. over a time period of several decades. Recently, /Brissonneau et al. 2004/ suggested that similar ageing processes may be of relevance also in materials to be used for long-term storage of nuclear waste. (Technical report TR-10-46, 2010)*

Based on the finding by Brissonneau et al. that a copper content of more than 0.05% in the cast iron is required for the phenomenon to be relevant, the process is neglected in the SR-Site. The microstructural damage to the copper shell is also neglected based on the study by Guinan:

*The specifications for the material for the cast iron insert require that the copper content be kept below 0.05%. The process is, therefore, neglected for the cast iron based on the results presented in /Brissonneau et al. 2004/. It is also neglected for the copper shell based on the results presented in /Guinan 2001/. (Technical report TR-10-46, 2010)*

Yet another issue identified in chapter two, the possible long-term radiation effects on the structure of the welded parts of the canister (refer to section 3.3.3 in chapter 2 of this study). This has not been addressed in SKB Process reports.
The structural damages possibly caused by neutron and gamma radiation to the engineered barriers is thus considered in the overall safety assessment procedure. However, the need of an investigation of the possible effects of direct and indirect radiation on the welded parts of the canister has not been addressed in the Process reports.

4.3.2 Bentonite microstructural effects

In the case of water-intrusion, termed as scenario 2 in chapter 2 of this study, the escaped α-emitting radionuclides may sorb onto the surface of the bentonite and cause radiation-induced microstructural damage to the bentonite buffer (refer to section 3.4.2 in chapter 2 of this study). This has been addressed in the TR-10-47:

*In the case of a failed canister, radionuclides sorbed in the buffer could possibly affect the properties by α-radiation.* (Technical report TR-10-47, 2010)

The issues are neglected in the SR-Site; as for the effects of gamma radiation on montmorillonite, it is argued in the report that the radiation doses need to be much higher than those expected for a radiation-induced transformation. In this regard, reference is given to a single experimental study by Pusch et al. (1993) where $^{60}$Co was used as gamma source to irradiate MX-80 bentonite samples. The samples received, according to the report, a total of $3 \times 10^7$ Gy and there was no evidence of significant change in montmorillonite quantity in either of the specimens. The expected dose is based on an earlier SKB radionuclide study (SR 97 /SKB 1999):

*In the radionuclide calculations for SR 97 /SKB 1999/, it was found that the total concentration of α-emitters in the buffer closest to the canister would give a total dose of $8 \times 10^{15}$ alphas/g of bentonite for the first million years with the assumption of an early canister failure. In most parts of the buffer, the α-dose will be much less since the α-emitters are strongly sorbed and will stay in the vicinity of the canister. $8 \times 10^{15}$ alphas/g corresponds to an absorbed dose of 8 MGy. /Gu et al. 2001/ show that the amorphisation dose for montmorillonite is 30 GGy. Since the total dose received is more than three orders magnitude lower than the amorphisation dose, the impact on the properties of the buffer can be expected to very small.*

Effects of radiation on the montmorillonite in the backfill is neglected in the SR-Site by referring to the same reason as the one for bentonite buffer i.e. too low dose rates for the radiation-induced decomposition of montmorillonite to be significant.

The radiation effects on bentonite buffer has thus been considered as a phenomenon by SKB. Effects of direct radiation has been mentioned and discussed.
4.3.3 Radiation effects on microbial activity

The effect of radiation on SRB has been briefly discussed in section 3.6 of chapter 2 of this study. As far as the coverage of the issue by SKB, the microbial processes are discussed in section 3.5.14 of Buffer and Backfill Process report (TR-10-47). Radiation intensity in terms of dose-rates and radiation-resistance of microbes has been mentioned as one of the relevant aspects for the process.

5 Radiation issues in future R&D plans

One of the purposes of the SR-Site project is to provide feedback to SKB’s RD&D (Research Demonstration and Development) programme. According to the SR-Site main report (Volume III, p. 837), general research on "processes of importance" should continue even though the status of the current knowledge is satisfactory:

In accordance with a proper safety culture, general research on processes of importance for safety should continue even if the current view is that existing knowledge is sufficient to demonstrate long-term safety. More specifically, there are some issues which SR-Site shows to contribute to risk and where the basis for the assessment can be improved through more R&D. (SR-Site, 2011)

5.1 Recommendations in SR-Site

A number of recommendations were made in SR-Site for further studies of the processes relevant for the functions of the engineered barriers and other aspects of the KBS-3 design. As for the fuel, it has been stressed that the research on fuel dissolution should continue. It is further stressed that research should focus on data from repository like conditions:

This research should focus both on data under repository like conditions and on understanding of basic processes contributing to fuel dissolution. Findings from this research could reduce the calculated risk further. (SR-Site, 2011)

As for the canister, the need for further studies of copper corrosion processes on the surface of the copper-shell in oxygen-gas free environment is stressed. In addition, deformation of cast iron and the copper canister are mentioned as the areas to be studied further. No specific mention of the need for further studies addressing direct or indirect radiation effects on the canister is found in the feedback for the future research and development.

As far the bentonite clay, further understanding of the transport in the buffer and backfill material is mentioned as an area for further research. Montmorillonite alteration and buffer erosion/colloid release have been mentioned as areas for further study. However, other aspects, rather than the effects of radiation on these phenomena, are pointed out as the areas where more understanding of the processes is needed.

5.2 Radiation issues in FUD programs

The need for further research and development after SR-Site (2011) have been discussed in Fud programs 2013 and 2016. Further plans for studies of issues mentioned in FUD-programs are
based on the discussions made in the FUD-programs. A summary of the discussions made in FUD-programs 2013 and 2016 and plans for future research of the issues relevant for this study are presented in the sections below.

5.2.1 Radiation induced dissolution of the fuel
Post 2011 research results concerning radiation induced dissolution of the spent fuel have been summarized under section 23.2.6 of FUD-program 2013. Results from experiments based on UO₂ doped with different species have been discussed. The role of Fe(II) and hydrogen formed due to anoxic corrosion of iron in consuming the radiolytically produced oxidants has been discussed by referring to studies by Cui et al. (2011).

By referring to a KTH study (by Nilsson and Jonson 2011), the findings concerning the catalytic decomposition of the radiolytically produced hydrogen peroxide on the surface of UO₂ are mentioned. The need for further understanding of the processes involved in radiation induced dissolution of the fuel has been emphasized.

Development in 2013 and afterwards in understanding of fuel chemistry and dissolution processes have been discussed under chapter 7.7 of the FUD-program 2016. Making references to open studies from 2013, the findings that fission products change the chemical properties of the UO₂ and its tendency to be oxidized are discussed. It is acknowledged that the extent of the influence of the fission products on UO₂ properties need to be investigated further.

By referring to a study by Lousada et al. (2013), the findings regarding the catalytic decomposition of H₂O₂, the main radiolytic oxidant, on the surfaces of UO₂ have been mentioned. The dependence of the H₂O₂ surface-decomposition behavior on the type of metal ions and the resulting influence on the kinetic parameters of the decomposition process have been mentioned. According to the report, the process of catalytic decomposition of H₂O₂ on the surface of the spent fuel may decrease the oxidative effects of the water-radiolysis products in the repository. A method described by Yang and Jonsson (2014) for detection of the OH-radicals formed on the surface of the metal oxides due to the catalytic decomposition of H₂O₂ has been mentioned. The overall reaction mechanism and a kinetic model for H₂O₂ decomposition on the surface of ZrO₂, developed on the basis of the method by the same authors (in 2015), are mentioned. The findings in 2016 by Barreiro Fidalgo et al. (2016) concerning the formation of H₂O₂ from H₂ and O₂ and catalyzed by ZrO₂ surface are mentioned.

According to the report, the research concerning the radiation-induced dissolution of the spent fuel will continue in collaboration with Royal Institute of Technology (KTH) in Sweden. The studies will, according to the report, concentrate on the processes that occur on the surface of the fuel and contribute to further understanding of the effects of the fission products on the radiation induced dissolution of the fuel. In addition, the effects of solid phase alterations and the composition of matrix on the kinetics and mechanisms of radiation induced dissolution of the spent fuel will be studied. The focus of the studies will be on UO₂ doped with gadolinium, neodymium and yttrium; however, effects of variations in particle size will also be included in the studies. These studies will be carried out within the framework of KTH PhD projects.
5.2.2 Cast iron and other ferrous material
The possible effects of gamma and neutron radiation on cast iron insert has briefly been mentioned in section 24.2.6 of the FUD-program 2013. More theoretical studies to understand copper precipitation phenomena was emphasized in FUD-program 2010. According to FUD-program 2013, calculations have been performed to examine how the production of defects in the matrix with the help of electron radiation could be used to study the mobility and precipitation of copper in iron matrix. There have however been difficulties in conducting radiation experiments due to technical reasons. According to the report, solubility and diffusivity of copper in iron will be studied further.

The issue of copper precipitation in cast iron has also been mentioned and discussed in FUD-program 2016. According to the report, the issue will be studied further to determine the maximum content of copper in the cast iron. In the future studies of the issue, the recent analysis (Toijer, 2014) resulting in new radiation levels expected at the surface of the cast iron will be used. Apart from the cast-iron insert, the studies will encompass the steel used in the fuel channels of the insert. In addition, the similar issue of Late Blooming Phases (LBP) in cast iron has been mentioned and discussed in the FUD-reports 2013 and 2016.

The possibility of radiation induced corrosion of cast iron has, however, not been discussed in the mentioned FUD-programs 2013 and 2016.

5.2.3 The copper shell
The findings in recent studies mainly by KTH concerning the radiation-induced corrosion of the copper shell have briefly been mentioned and discussed in section 3.3.1 of chapter two of this study. As far SKB’s covering of this subject, the FUD-program s 2013 and 2016 have addressed these issues in some detail. In Fud program 2016, the studies by Björkbacka et al (2013 and 2015) within the framework of KTH PhD projects have been referred to. It has been specifically pointed out that the dose rates used in the experiments by Björkbacka are much higher than those expected on the surface of the copper in the repository. It is, however, acknowledged that the exposure times have been adjusted so that the total dose received by the copper shell will not be far from those expected at the surface of the copper shell.

According to the report, the results of radiation-induced corrosion of the copper shell by Björkbacka et al. indicates that radiation-induced corrosion within the doses expected in the repository will be lower than those calculated (pessimistically) in the SR-Site. Reference has also been made to a study by Ibrahim et al. (2015) within the Canadian nuclear waste program in which copper corrosion is studied under radiation field. The radiation dose (0.35 Gy/h) used in the experiments by Ibrahim et al. are not far from the dose expected at the surface of the copper in the KBS-3 design. Experiments were also carried out under much higher (10 000 times) radiation doses which indicated higher corrosion i.e. a layer thickness in the order of < 1 µm was observed. According to a comment by the SKB, the experiments under higher doses (than those expected in the repository) may be of some value for understanding the
mechanisms; however, extrapolating the results to the lower doses during a long period may not be appropriate without a detailed knowledge of the involved mechanisms.

According to the FUD-program 2013, the PhD project at KTH studying the effect of gamma radiation on copper corrosion would continue; the role of dose and dose rates would also be examined. According to the FUD-program 2016, KTH will continue the research on reaction mechanisms to understand the effect of oxide layer on radiation induced corrosion of copper in groundwater environment expected in the KBS-3 repository. SKB will also follow the work carried out concerning this type of corrosion within the Canadian nuclear waste program.

As for the radiation-induced copper precipitation, the need to study the extent of the problem in order to be able to assess the maximum copper content allowed in the insert cast iron material is acknowledged in the report. SKB refers to an experimental study carried out by the department of Reactor Physics at KTH concerning the Cu-precipitation issue. The study shows, according to the report, that the effects of the radiation on the cast iron samples provided by SKB, are insignificantly low.

According to the SKB, the company plans to continue studying the issue of copper precipitation to ensure that the requirements regarding the copper content in the cast iron for the insert are appropriate. In addition, the effect of radiation on the insert steel channels for the fuel will be included in the studies. However, the details regarding the focus and the extent of the studies have not been provided in the report.

The effects of radiation on the FSW joints (welded parts) have not been discussed in the report. However, according the report, the material properties of the welded parts are comparable to those of the original material (the bulk copper in the canister).

Since the atmosphere in the canister will be replaced by argon (refer to section 3.2.4 of chapter two of this study), there has been a need for studies to investigate the possibility of the radiation-induced corrosion of the ferrous metals and copper under argon atmosphere. As studies addressing the issue have almost been non-existent, there have been no mention of this phenomenon in the FUD-programs.

5.2.4 The buffer and the backfill
The radiation effects on the microstructure of the bentonite in terms of radiation induced structural decomposition is mentioned very briefly within a relatively large section in FUD program-2016 about a number of processes affecting the buffer properties. According to the report, bentonite may be decomposed by ionizing radiation, but the radiation doses expected at the surface of the buffer will be too low to cause any significant structural alterations. No mention of any studies covering the subject has been made in the report. However, in FUD-program 2010, the effects of direct radiation on the microstructure of bentonite has been declared to be insignificant due to very low doses. According to the report (FUD program 2010), handling of this issue in SR-Site project will be based on the same data used in safety assessment carried out by the French national radioactive waste management agency (ANDRA -Dossier-2005). It is
stated in the report that no further studies concerning this issue were planned. As for the indirect effects of radiation (via radiolysis of water), the FUD-2010 program stated that the thickness of the copper shell is enough to ensure that the gamma radiolysis will be neglectable outside the canister.

6 Discussion

Most of the issues mentioned in chapter 2 of this study have either been mentioned and discussed in the SR-Site, the underlying process reports or other SKB technical documents. The issues discussed after SR-Site submission in 2011 and FUD-program 2010 reflects well, albeit with some exceptions, the most significant developments in terms of findings and observations reported in the open scientific literature. The thoroughness of the SKB’s work in some cases relevant to the subject of this study can, however, be debated.

Most of conclusions in SR-Site and the underlying process reports related to radiation induced processes are mostly based on the type and level of the radiation from the SNF in terms of dose rates the engineered barriers will be exposed to. Due to the difficulties associated with conducting experimental studies involving high radiation doses, the values of dose rates have in most cases been based on calculations and estimations. This is the case both for the study by Håkansson on gamma and neutron fluxes and the processes concerning the issue of radiation attenuation. As there may be differences in the results of such calculations, it probably is advisable to avoid depending altogether on the dose rates and accumulated doses while assessing the relevance of significance of radiation induced processes in the KBS-3 repository design.

As there may be uncertainties, albeit minor, associated with values of the decay data (Helmer & Chechev, 2017) (Browne, Bé, MacMahon, & Helmer, 2001), it may be argued that several uncertainties in a process where evaluations are based on other evaluations may lead to some significance in the associated uncertainties.

Radiation doses outside the canister were recalculated by Klas Lundgren (TR-04-06) as a result of discussions concerning the accurateness of the first calculation. Even though the results of studies do not differ much in terms of dose rates outside the canister, such discussions may be seen as an indication of uncertainties associated with conclusions based on such calculations. Thus, the evaluation of significance of the relevant radiation related processes should be carried out on the basis of thorough understanding of the relevant processes regardless of the radiation levels.

The need for thorough understanding of the processes, referred to by SKB as ökad processförståelse, relevant for the safety of the KBS-3 repository concept, has, however, been acknowledged by the company. One of the purposes of SKB’s research and development program is to contribute to an enhanced understanding of such processes. Several radiation related processes that have been neglected in SR-Site, mostly due to the expected (calculated) low radiation doses, have been given due importance in the FUD-programs. Based on the information in SKB-documents summarized in the sections above, a discussion of the thoroughness of SKB work in each case is presented in the lines below.
The fuel dissolution

As for the safety analysis of the fuel in terms of radiation-induced oxidative dissolution, most of the significant research developments have been mentioned and discussed in the FUD programs 2010-2016. Even though there have been numerous studies addressing the subject, prediction of release rate of the radionuclides due to radiation-induced dissolution requires detailed understanding of a number of factors affecting the dissolution rate of the fuel matrix. These factors include geometrical dose distribution, the radiation chemistry of the aqueous phase and the effects of the groundwater composition. As far reactions taking place at the surface of the fuel, substantial knowledge of the kinetics of oxidation/reduction, adsorption, dissolution and diffusion is required (Jonsson, 2012). The surface reactions have been identified by SKB as an area where further research is required. The planned KTH PhD studies will also focus on effects of solid phase alterations on radiation-induced dissolution of the SNF and particle size variations (refer to the relevant sections above). Even though SKB’s identification of the most relevant issues in this regard can be appreciated, it is evident that a limited number of PhD projects may not be enough to cover all the research needs identified.

Ferrous materials

Studies covering the long-term effects of radiation, in terms of radiation induced corrosion and microstructural defects, on nodular cast-iron of the type chosen for the KBS-3 repository are limited in numbers. As for the microstructural effects of radiation on the fuel cladding and the insert fuel channel material (zirconium alloy and steel, respectively) in the fuel, the short-term effects of high radiation doses are well-known due to exposure of these materials to radiation during reactor operations. The majority of the studies addressing the long-term effects of radiation on these materials, including the relatively recent master thesis (Toijer, 2014) (Messina, 2010), have been based on theoretical calculations of doses and dose rates. These studies help a great deal in shedding light on this very important subject. These calculations include the most pessimistic scenarios in terms of radiation doses and the results still indicate insignificant effects in terms of microstructural effects. Thus, there may be little reason to argue for more theoretic studies of the radiation effects on the ferrous materials that will be exposed to ionizing radiation from the SNF. As for the experimental studies addressing the subject, there may be uncertainties associated with extrapolating the known effects of high dose rates under shorter periods to long-term effects under low doses. More experimental laboratory scale studies of long-term radiation effects on ferrous materials may contribute to removal of the remaining uncertainties. This is especially true about the insert material (nodular cast iron) whose response to both short- and long-term radiation effects are poorly studied under the repository conditions. It is, off course, not possible to reproduce the exact conditions expected in the repository and the length of the repository time scales. However, considering the very limited number of such studies, it may be advisable to allow more resources for laboratory-scale experimental studies of long-term effects of radiation on these materials.

As for the radiation-induced corrosion, there is a need for a better understanding of how cast-iron of the specific type planned for the KBS-3 repository will respond to radiation in the long-term. One may argue that the expected radiation doses at the surface of the insert are too low and/or the hydrogen gas produced due to corrosion of the ferrous materials may limit the amount of radiolytically produced oxidants. Studies covering this subject should, however, investigate the
possibility of radiation-induced corrosion regardless of the radian levels expected at the surface of the insert. This is in line with SKB’s efforts in terms of a better understanding of the processes (uttökad processförståelse) relevant for the long-term safety of the KBS-3 repository. There is a certain value in conducting experimental studies of the specific type of the cast-iron rather than using findings from the studies of other parts of the system to discuss the possibility of radiation-induced corrosion of the insert.

Studies addressing the issue of radiation induced corrosion of cast iron should obviously encompass other ferrous materials in the system, such as the steel in the channel for fuel in the insert.

*Copper precipitation*

Copper precipitation phenomena in nuclear reactors is a well-understood subject. However, as in the case for radiation effects on cast-iron, the long-term effects of radiation on cast iron in terms of copper precipitation needs to be studied experimentally. SKB motivates the adequacy of references for each process mentioned and discussed the SR-Site report. As for the study by Brissonneau et al. (2004), the fact that the study has been based on low-carbon steel, rather cast iron, has been mentioned in the SR-Site main report. As for the study by Guinan in 2001, it is acknowledged that the report is not a peer- or factual reviewed, but "scientifically consistent with other references that are peer reviewed articles or papers". Irrespective of the adequacy of the references in terms of peer- and factual review, it may be advisable to consider the number of the studies concerning the subject. There should be a reasonable number of studies covering or addressing the issue in order for the decision of neglecting to be well-founded.

In the assessment regarding the adequacy of the study by Guinan et al (2001), it is stated in SR-Site main report that the study is made directly for the KBS-3 canisters. However, in the study by Guinan, pure iron rather than the cast iron produced through a different manufacturing process for the insert, was assumed to be the material. This has been acknowledged in the Fuel and Canister Process report; an explanation or scientific arguments as to why this is not considered is not offered in the report.

There have lately been more studies concerning the phenomena. A KTH master degree project report supports the earlier findings (Toijer, 2014). The study presented a new model for damage flux in the cast iron arguing that the earlier models have been questioned. The results based on the new model indicated that earlier damage flux may have been overestimated. As for the experimental studies, a rather recent study by Olsson et. Al. (Electron irradiation accelerated Cu-precipitation experiment) on the effects of gamma radiation on the Cu-precipitation (Bilaga R-13-50) reported insignificant effects even in the long-term perspective. Cast iron samples were used in the study and an accelerated ageing experiment through electron irradiation, mimicking gamma radiation, was carried out. The sample was irradiated throughout a week, corresponding to 45 years in the repository, according to the acceleration factor of the ageing processes calculated in the study.

The studies may have been few in numbers; however, the outcomes of the studies have been the same i.e. Cu-precipitation may not occur under repository conditions even in the long-term
perspective. Brissonneau et al. who were the first ones to discuss the possibility of this phenomenon under the repository conditions, recommended experimental studies (irradiation experiments) to confirm the results they acquired. The investigation by Olsson et al., a SKB study, is well in line with the recommendations.

Copper shell
Radiation induced corrosion of the copper shell has been mentioned and discussed in both the SR-Site main report and the FUD-program reports. As in the case of other processes, low radiation doses are mentioned as the cause of neglecting the process in SR-Site. The main SKB technical report covering corrosion processes in the copper shell is Copper Corrosion Under Expected Conditions in a deep geological repository (TR-10-67) which was published in 2002 and updated in 2010. As for the radiolysis effects on the copper corrosion processes, the Fuel and Canister Process report bases much of its information on TR-10-67. In the report, an assessment regarding the possibility of enhanced corrosion due to water radiolysis, by referring to a number of studies, is made according to which there is no evidence for enhanced corrosion rates due to gamma radiation. The radiation induced corrosion may, according to the report, be possible at very high doses (> 100 Gy/h), but at lower doses, the radiation may rather lower the corrosion rate.

The observations made in the studies by KTH differ fundamentally from those mentioned in the report (Björkbacka, Hosseinpour, Leygraf, & Jonsson, 2012) (Björkbacka, Johnson, Leygraf, & Jonsson, 2017). Significant corrosion rates of copper shell as a result of gamma radiation have been reported and discussed in chapter two of this study. The cumulative radiation doses are within the range of those expected at the surface of the copper. These observations have, however, been mentioned and discussed in FUD-program 2016 (refer to the relevant sections above). The findings by Björkbacka et al. concerning the radiation-induced formation of Cu$_2$O and the local cavities formed on the surface of the copper samples have been discussed in the report. By refereeing to the study by Björkbacka in 2014 (Björkbacka, 2014), the observed higher amount of oxidized copper in case of an initially existing oxide layer has been mentioned.

SKB acknowledges the need for more studies to understand the mechanisms involved in radiation-induced corrosion of copper under anoxic conditions. As has been mentioned in the relevant sections above, SKB’s collaboration with KTH in this regard will continue, according to the FUD-program 2016. The role of oxide film in the radiation-induced corrosion of copper in groundwater under repository conditions is specifically mentioned as an area where further research will be carried out at KTH. However, as in the case of the radiation-induced fuel dissolution, more research efforts may be required to investigate the mechanisms involved in this process.

Most of the studies referred to in the section (7.3) of TR-10-67 dealing with corrosion of copper in water in the presence of gamma-radiation were carried out in 1980's and 1990's. These rather old studies generally do not thoroughly reflect the conditions expected in the Swedish concept of the repository (KBS-3). The results of a comparatively recent study conducted in 2003 (Corbel et al.) referred to in the section of the of TR-10-67 indicated higher corrosion risks as compared to the ones carried out earlier. The most recent in this series of studies is the ones carried out by
Björbacka et al, which, as in the case of the study by Corbel et al. indicated significant corrosion of the copper under anoxic conditions.

There is also a general lack of clarity in discussions surrounding the effects of dose rate. There seems to be discrepancies in results of the older and newer studies in terms of effect of dose rates, especially the possible/potential effects of long-term radiation (repository time-scales) with low dose rates.

The fundamental difference in results of the older and more recent studies in terms of the extent of radiation induced corrosion of copper under reducing conditions calls for a general renewal of understanding of these mechanisms. The role of the dose-rate needs to be discussed further.

**FSW joints**
According to the Fuel and Canister Process report and the SR-Site main report (Technical report TR-10-46, 2010) (SR-Site, 2011), the effects of chemical and mechanical conditions in terms of corrosion of the welded parts are of no importance in the long-term. In this regard corrosion caused by earth currents, by deposition of salt on the copper surface and SSC has been mentioned as negligible. The effects of radiation induced corrosion of FSW joints has not been specifically discussed. To the knowledge of the author, the possible effects of direct radiation or radiation induced radiolysis of water on the welded parts of the copper shell has not been discussed elsewhere either. As mentioned in chapter two of this study, the resulting weld may differ from each other in terms of mechanical properties depending on a number of factors, such as the speed of the tool, axial-force variation, applied load and the position of the probe tip. There is thus a certain possibility that FSW joints may respond differently to radiation as compared to the bulk material (the copper shell). Separate experimental studies addressing radiation induced corrosion and microstructural alterations of FSW joints are therefore important to be conducted.

**Bentonite buffer**
The effects of gamma and alpha radiation on the montmorillonite in terms of microstructural alterations and the indirect effects (via water radiolysis products) have been neglected in SR-Site (refer to the relevant sections above). No further research for understanding of the radiation-induced processes affecting the bentonite clay are planned by SKB. Some important research activity concerning this issue has been ongoing at KTH since FUD-program 2010 and earlier. According to one such study from 2012, the structural Fe(II)/Fe_{Total} ratio (refer to section 3.4.1 of chapter 2 of this study) of montmorillonite is increased as an effect of gamma radiation under anaerobic conditions. In addition, a strong co-relation between the structural Fe(II)/Fe_{Total} ratio and the H_{2}O_{2} decomposition rate in montmorillonite dispersions was observed (Holmboe*, Mats, & Susanna, 2012). These findings are relevant for the safety analysis of the bentonite buffer since the results of the study indicate that the structural iron in montmorillonite may function as a sink for H_{2}O_{2}, the main radiolytically produced oxidant.

In an earlier study (2009) by the same authors, significant radiation effects on stability of the colloids (discussed under section 3.5.2 of chapter 2 of this study) have been observed. (Holmboe, Wold, Jonsson, & Garcia-Garcia, 2009). The dose rate used (0.154 Gy/s) and the total doses
received (26.6 kGy) the MX-80 bentonite sample within 48 hours is not far from the levels that may be expected at the surface of the bentonite buffer in the repository.

The developments in understanding of the mechanism involved in the radiation-induced of montmorillonite has not been mentioned/discussed in FUD-programs since 2010. As for the handling in SR-Site, the assessment is based on data used in the safety assessment carried out by ANDRA. A thorough analysis of research by ANDRA in this regard is out of the scope of this study. However, the available open literature information, such as those mentioned above, are indicative of the need for more work to understand the mechanisms involved in the radiation induced effects on montmorillonite and its colloidal dispersions.

The purpose of Long-term tests (LOT) of bentonite buffer, mentioned in chapter 1 of this study, is to investigate model and hypotheses of the physical properties of the buffer material. Apart from the properties of the buffer, long-term corrosion of copper, for example by bacterial activity, is studied. To simulate the effects of temperature from the radioactive decay of the SNF, heaters are used (refer to chapter one). The heaters may replicate the effects of the temperature, but long term effects of radiation induced processes, such as effects of radiolysis processes, are not studied by these tests. Contrary to radiation-induced corrosion of copper in which the damage is assessed in terms of the depth of the corroded area, it is the possible changes in material properties of the buffer which is the important aspect in terms of long-term safety of the repository. As the buffer stability directly effects the corrosion possibility of the copper canister in the KBS-3 design and mobility of the escaped nuclides, investigating issues that may affect its properties under the conditions more specific for KBS-3 design, rather than other similar programs, should be a priority.

The effects of radiation on bacterial activity in the montmorillonite pore water has briefly been discussed in Buffer, Backfill and Closure report. According to report, microbes are usually resistant to radiation, but this may depend on the type of microbe. Increasing radiation doses leads to decrease in the numbers of surviving species. To the knowledge of the author, there has been no studies indicating the possibility of an increase of the number or of the activity of the microbes due to radiation. However, the possible results of long-term radiation on the bacterial activity specifically under conditions expected in KBS-3 repository have yet to be demonstrated.

Metal-metal interfaces
The possible effects of radiation on the processes at the metal-metal interfaces (mainly cast-iron interface) and metal-clay (copper-bentonite) interface have not be studied as a subject per se. The possible processes taking places at the copper-bentonite (MX-80) interface have been discussed in terms of bentonite interaction with metal corrosion products within other nuclear waste repository concepts (Sellin & Leupin, 2014). How these processes may be affected by the radiation from the SNF has not been studied to the knowledge of the author of this report. As the redox conditions govern corrosion processes, the possible effects of radiation on the corrosion mechanisms, in case of water intrusion, at the insert-copper interface should specifically be studied.

Copper corrosion in argon atmosphere
The possibility of radiation-induced corrosion of copper in argon atmosphere has recently been
studied at KTH (Björkbacka, Radiation induced corrosion of copper, 2014)(Björkacka, Johnson, Leygraf, & Jonsson, 2017). As the findings (refer to section 3.4.2 of chapter 2 of this study) i.e. the possibility of significant corrosion under argon atmosphere is very relevant for the safety analysis of the copper canister, research into the phenomenon should continue.

The study also addresses the issue of radiation-induced corrosion of copper in humid air atmosphere. The corrosion behavior of copper was found to be similar in both cases (argon and humid air atmospheres), which indicates that it is the hydroxyl radical, rather than HNO₃, responsible for the observed corrosion of the copper.

These findings were reported in 2015, the FUD-program 2016 is thus expected to mention and discuss the results of these studies. The findings in this study and those covering radiation induced effects on montmorillonite are indications of a need for a more proactive approach in terms of studying the mechanisms of radiation-induced phenomena affecting the engineered barriers of KBS-3 repository design.

**Unknown unknowns**

In many cases, the now known effects of the radiation on materials were once unknown to exist, for example the case of copper precipitation in iron and the radiation-induced activation of corrosion products deposited as crud in the nuclear reactors. Many of these phenomena were discovered as the result of radiation of materials during the reaction operation. It is thus of utmost important to be prepared for new observations (factor whose existence have not come to light yet) associated with long-term radiation of materials relevant for the safety analysis of the engineered barriers. For example, the cast iron insert has a number of other metals, such as nickel, manganese and chromium, within its structure and the copper shell has several metals including Ag. There may be unknown factors associated with how these materials respond to long-term radiation from the SNF under the repository conditions both in scenario 1 and scenario 2. To unveil or discover such unknown unknowns, it is important that the studies addressing radiation issues may be carried out under conditions as close to those expected in the repository over different periods of time as possible. As for the reactor materials, the effects of high doses are well-known due to the knowledge accumulated over the decades of nuclear reactor operations world-wide. On the contrary, knowledge related to exposure of cast-iron of the specific type for the KBS-3 and the oxygen free copper of the type used for the canister to different kind of radiation from the SNF over a very long period is very limited. In addition, long-term exposure of bentonite clay under repository conditions is a rather new phenomenon and, thus far, a poorly studied subject.

7  **Conclusions**

Most of the relevant aspects of the effects of radiation-related processes on the engineered barriers within the KBS-3 repository design have been considered in SKB's overall safety assessment procedure. The majority of the radiation induced processes relevant for the concerned safety assessments have, however, been neglected in the safety assessment SR-Site. Neglecting these processes in SR-Site are motivated mainly by the results in SKB's radiation evaluation and
attenuation studies according to which doses/dose rates maybe too low to cause significant water radiolysis or structural damages to the engineered barriers.

The developments in understanding of the mechanisms involved in the radiation induced processes have been mentioned and discussed in the FUD-program reports. Most of the research developments discussed in the FUD-programs reflect the scientific findings with the exception of some aspects. These aspects include radiation effects on montmorillonite which has not been discussed in SKB documents to the extent needed considering the importance of the issue. The developments in this regard by the studies carried out at KTH should be given due importance in the future FUD-programs. Another issue not discussed in SKB documents is the radiation effects on the welded parts of the copper shell. Experimental studies need to be carried out to investigate the possible different response of the welded parts to long-term radiation, both in scenario 1 and scenario 2.

The findings mentioned in the FUD-programs regarding the issue of radiation-induced dissolution of SNF are in line with the developments. The need for further research has been acknowledged and the collaboration with KTH in this regard will continue. However, the issue may require more research efforts than those planned.

As for the radiation effects on the cast iron, there probably have been enough theoretical studies to investigate the radiation-induced microstructural effects. More experimental studies are advisable to remove any remaining uncertainties in this regard. The possibility of radiation-induced corrosion of the cast-iron may require more research. The research efforts put into the issue of copper precipitation phenomenon in cast-iron may be satisfactory.

Both the direct effects of radiation and indirect (via radiolysis of water) on copper has been mentioned and discussed in SR-Site and the FUD-programs. However, the findings mentioned by SKB in SR-Site and the observations made in the recent studies by KTH differ a great deal in terms of the extent of radiation-induced mechanism. There may be a need to carry out studies for an update on the status of the understanding of the involved mechanisms. A clarity regarding the role of the dose rate needs more discussions and probably more studies.

Corrosion of copper and ferrous materials under argon atmosphere has not been mentioned or discussed as a phenomenon in SKB documents identified as relevant for the subject of this study. The recent findings by KTH in this regard needs to be presented and discussed in FUD-programs including the plans for the future research.
8 References


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