

## **SKB TR-11-01**

# **Long-term safety for the final repository for spent nuclear fuel at Forsmark**

## **Main report of the SR-Site project**

### **Volume III**

In the earlier distributed report, there are errors that have now been corrected. The corrected pages 664, 665, 666, 723, 730, 846 and 847 are enclosed. The changed text is marked with a vertical line in the page margin. An updated pdf version of the report, dated 2012-12, can be found at [www.skb.se/publications](http://www.skb.se/publications).

The changes on pages 664-666 are due to an error in the input data for the earlier calculation of clay assisted fuel dissolution. With corrected input data, an extended analysis and discussion is required compared to that in the earlier version, hence the added text and the updated figure on these pages. The new material has been factually reviewed according to the routines for SR-Site reports. As seen in the text, the conclusion regarding clay assisted fuel dissolution is unchanged.

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## Chemical influence of major and minor groundwater components

*Range in which the fuel dissolution rate is valid according to the Data report:*  $\text{HCO}_3^- < 0.01 \text{ M}$ . The beneficial effect of molecular hydrogen in homogeneous radiolysis is based on its reaction with the OH-radical, which converts the very oxidising radical ( $\text{OH}\cdot$ ) to water and a very reducing radical ( $\text{H}\cdot$ ). Bromide ions are strong reductants and react ~250 times faster with the OH-radical than the  $\text{H}_2$  molecule. It is said that bromide scavenges the OH-radical, thus decreasing the beneficial effect of hydrogen on homogeneous water radiolysis. The radical-rich beta and gamma radiations are expected to have decayed to negligible levels at the time of canister failure in the corrosion scenario. This together with the absence of very saline waters during repository evolution and the very low alpha activity makes any influence of bromide on fuel dissolution very improbable.

*Ranges in evolution cases studied:* The carbonate concentrations during the whole repository evolution are expected to be below 0.01 M.

*Need for analyses of additional evolutions to rule out unfavourable conditions:* The total concentration of inorganic carbon, which at pH values close to neutral is close to the concentration of  $\text{HCO}_3^-$ , is highest in the upper parts of the rock where recharge of meteoric waters dominates, because several biological processes produce  $\text{CO}_2$  in the soil. In deep saline waters, rich in  $\text{Ca}^{2+}$ , the concentration of carbonate is kept low because of equilibrium with calcite. Typical carbonate concentrations for waters at Forsmark are illustrated in Figure 10-45. Thus, the only process that can increase bicarbonate concentrations in groundwaters is the enhanced infiltration of fresh meteoric waters. The data at Forsmark shows that the waters having a large influence of meteoric recharge all have carbonate concentrations around or below 0.01 M, and it is therefore concluded that there is no need to analyse the fuel dissolution rate for higher total carbonate concentrations.

## Additional factor identified: Transport of uranium with clay colloids in the groundwater

When the buffer is partially or completely eroded, a cavity filled with a slurry (gel/sol) of water containing colloidal clay particles may exist in the deposition hole. Should the canister be breached under such circumstances, the clay particles themselves are not expected to affect the oxidative fuel dissolution rate. Dissolved U(IV) would, however, be expected to sorb strongly to the clay particles. This sorption increases the amount of U(IV) released into solution from the re-precipitated  $\text{UO}_2(\text{s})$  or the fuel matrix. In this case, the amount of U(IV) sorbed on clay particles may be calculated as the  $K_d$  value for U(IV) on clay particles multiplied by the U(IV) concentration in solution, determined by  $\text{UO}_2(\text{s})$  solubility. No limit to the U(IV) release rate from  $\text{UO}_2(\text{s})$  to satisfy U(IV) solubility limits in the canister void is then posed. In the case when all re-precipitated  $\text{UO}_2(\text{s})$  is dissolved due to sorption to clay particles, the remaining U(IV) needed to saturate clay particles is released from the fuel matrix, resulting in an increase of the fuel dissolution rate. To calculate this enhanced U release, the values for  $\text{UO}_2(\text{s})$  solubility and  $K_d$  for U(IV) on clay particles are taken from the **Data report**. The rate at which the clay particles with sorbed uranium are carried away are set by the concentration of clay particles and water flow rate. These depend on the scenario analysed.

*Need for studies of additional evolutions to rule out unfavourable conditions:* Fuel dissolution in the presence of a clay slurry needs to be considered. This is done as described below.

The eroded void volume of the deposition hole is assumed to be filled with clay particles that enhance the dissolution rate of the  $\text{UO}_2$  fuel matrix. The outward transport rate of U,  $R_U$  (mole/yr), is then obtained as

$$R_U = C_{\text{Sol}}U \cdot q \cdot (1 + C_{\text{Clay}} \cdot K_d)$$

where

$C_{\text{Sol}}U$  is the solubility of U(IV) (mole/m<sup>3</sup>),

$q$  is the advective flow at the deposition hole (m<sup>3</sup>/yr),

$C_{\text{Clay}}$  is the concentration of clay in the flowing fluid (kg/m<sup>3</sup>),

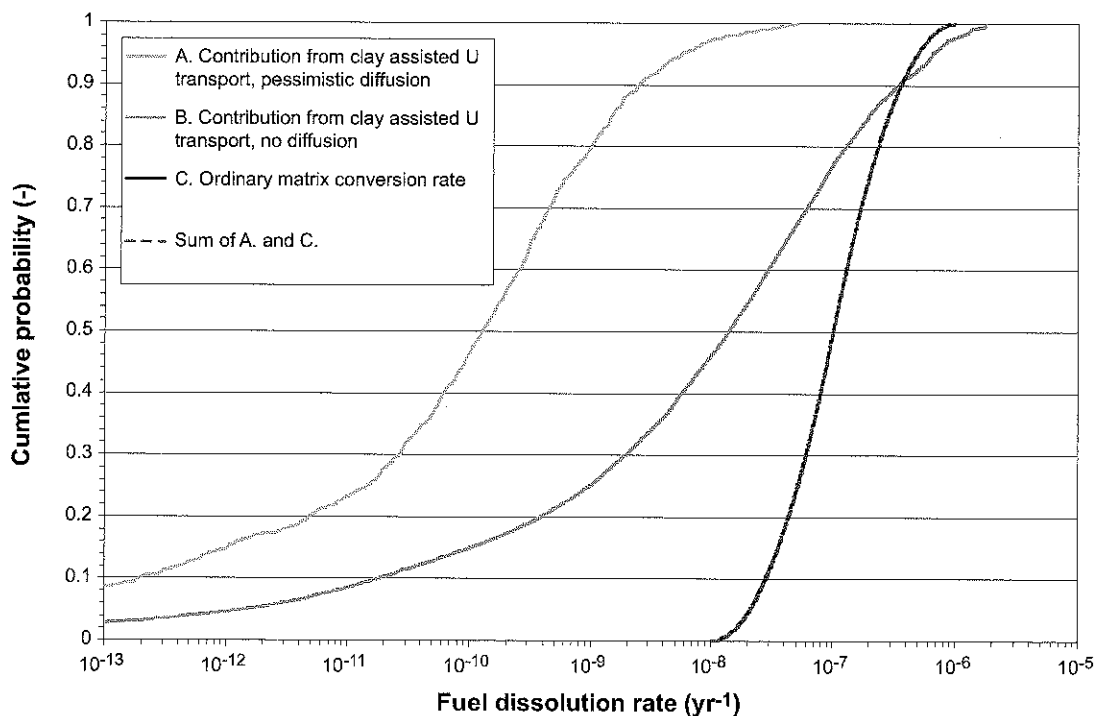
$K_d$  is the partitioning coefficient between solid phase and solution for U in a clay slurry (m<sup>3</sup>/kg).

In /Birgersson et al. 2009/ the void ratio when the transport of bentonite is by colloid dispersion is determined to be 73, which corresponds to a dry density of 37.6 kg/m<sup>3</sup>. This concentration will not be maintained in a large void in the deposition hole, since there will be a concentration gradient from the source of the colloids to the fuel. The location of the source could be remaining bentonite in the deposition hole, the backfill in the tunnel or through a fracture from another deposition hole/tunnel in the repository. Since it is impossible to determine the location of the source, and it may change with time, the value for  $C_{\text{Clay}}$  cannot be calculated. Instead a fixed value of 10 kg/m<sup>3</sup> is selected and judged to be a reasonable estimate of an upper bound of the mean value taken over the entire void volume.

Using  $C_{\text{Clay}} = 10 \text{ kg/m}^3$  and distributions of  $K_d$ ,  $C_{\text{Soil}U}$  and  $q$  for the central corrosion case, the contribution from U transport on clay particles to the fuel dissolution rate is calculated probabilistically and compared to the probabilistically sampled ordinary fuel dissolution rate for the central corrosion case.

The following also applies in the modelling.

- This process is only active during the relatively limited periods when the deposition position is exposed to dilute groundwater such that clay colloids can form. This is not taken into account in the calculation.
- Inflowing groundwater may have U concentrations exceeding  $10^{-9} \text{ mol/L}$ , but the influence of the occupation of sorption sites of clay particles by natural uranium present in the groundwater is pessimistically neglected.
- Should sorption to clay colloids be modelled as irreversible and assuming that all sorption sites were to be occupied by U, then a transport rate of the order of  $2.5 \cdot 10^{-3} \text{ moles/yr}$  is calculated for a sorption site density of  $2.0 \cdot 10^{-6} \text{ moles/g}$  /Bradbury and Baeyens, 2005/, cited in /Ochs and Talerico 2004/,  $q = 100 \text{ L/yr}$ , and  $C_{\text{Clay}} = 10 \text{ g/L}$ . This, in turn, corresponds to a fuel dissolution rate of approximately  $3 \cdot 10^{-7} \text{ /yr}$ , i.e. slightly lower than the maximum value of the “ordinary” distribution. An upper limit on the transport rate determined by the product of the sorption site density,  $q$ , and  $C_{\text{Clay}}$  is imposed in the calculation of the distribution in Figure 13-20, curve B. (Irreversible sorption, as well as U occupancy of all available sites is, however, ruled out for the conditions in the deposition hole, based on the description of the sorption process in the **Buffer, backfill and closure process report**.)
- Although advective conditions are assumed to prevail in the deposition hole, there will be parts of the path between the inner parts of the, assumed damaged, fuel elements and the canister exterior where the transport is diffusion controlled. A certain continuous outward, clay assisted transport of  $\text{UO}_2$  then requires a corresponding flux of  $\text{UO}_2$  over the diffusion controlled path. The geometry for this transport is difficult to assess, but an upper bound can be estimated as follows: Assume a pessimistically high cross sectional area of  $0.1 \text{ m}^2$  and a pessimistically low diffusion path length of  $1 \text{ mm}$ . Taking then the highest U solubility in the distribution used in SR-Site, i.e.  $4.2 \cdot 10^{-4} \text{ mol/m}^3$ , and assuming a U diffusivity in water (at  $11^\circ\text{C}$ ) of  $0.01 \text{ m}^2/\text{yr}$ , one obtains a maximum U flux of  $4.2 \cdot 10^{-4} \text{ mol/yr}$ , assuming a zero concentration  $1 \text{ mm}$  away from the fuel. With  $8,400 \text{ mol U/canister}$  this corresponds to a fractional dissolution rate of approximately  $5 \cdot 10^{-8} \text{ /yr}$ . This pessimistic bound suggests that the effect of U transport with clay particles is negligible in the radionuclide transport calculations. A bound using these diffusion data in combination with the distribution of U solubility is imposed in curve A in Figure 13-20.



**Figure 13-20.** Cumulative distributions of fuel dissolution rate. The contribution from clay assisted U transport has a negligible impact on the distribution for the ordinary matrix conversion rate. See text for further explanations.

As seen in Figure 13-20, the clay assisted fuel dissolution rate (curve B) is considerably lower than the “ordinary” dissolution rate (curve C) up to the 90<sup>th</sup> percentile of the distribution, whereas it is comparable or slightly higher above the 90<sup>th</sup> percentile. If also the pessimistic diffusion limitation is imposed (curve A), the clay assisted dissolution rate is well below the “ordinary” dissolution rate over the entire range. Therefore, it is concluded that the contribution to fuel dissolution from U transport is negligible.

### **Intrinsic conditions**

The following intrinsic conditions/processes have been considered in the **Fuel and canister process report** which provided the bases for the derivation of fuel dissolution data in the **Data report**: Alpha self irradiation, enhanced diffusion (ASIED), helium build-up and the influence of high burn-up.

### **Conclusion**

Based on the above account, it is concluded that the distribution of fuel dissolution rates given in the **Data report** is valid for all evolutions that need to be considered in SR-Site, even for groundwater conditions in which a clay slurry may contribute to the outward transport of uranium.

### **Metal corrosion rate**

*Range in which metal corrosion release rates are valid:* The release rates of the activation products from the metallic parts given in the **Data report** are based on a range of corrosion rates for stainless steel under anoxic conditions. These corrosion rates were estimated from a number of relevant studies carried out at temperatures up to 140°C and salinities up to anoxic sea water. The important parameters are thus salinity (especially chloride concentration) and redox conditions.

*Ranges in evolution cases studied:* In the corrosion scenario, canister failure occur after typically hundreds of thousands of years. In such time frames, the fuel temperature is close to the background temperature of the host rock, i.e. no increase of corrosion rates due to high temperatures is expected. The highest chloride concentrations expected during the whole repository evolution time are of the order 0.35 mol/L, i.e. much lower than in anoxic sea water. Hence the corrosion rates used in the **Data report** are valid (with a large margin) for the corrosion scenario.

*Need for analyses of additional evolutions:* The important parameters for the metal corrosion rate are the chloride concentrations and the redox conditions. As stated in the previous section about fuel dissolution, the analysis of the reference evolution includes the highest salinities that can be expected at Forsmark. The analysis of the redox conditions for the reference evolution shows a variability of redox potentials, always with anoxic conditions. Only the penetration of oxygen rich glacial melt waters can possibly lead to a deviation, and as described in Section 10.4.7 this is a very low probability event that is deemed to be possible only if in addition a set of extreme assumptions are made. Even if extreme assumptions were to be made, and oxygen postulated to reach a breached canister, the large reducing capacity of the remains of the failed copper canister and of the iron insert, would imply that anoxic conditions would prevail inside the canister. It is therefore concluded that there are no additional evolutions that need to be assessed.

### **Rock porosity, rock diffusivities and rock sorption coefficients**

#### **Rock porosity and effective diffusivity**

The rock porosity and effective diffusivity provided in the **Data report** (Section 6.8) are valid for all conditions in the host rock during repository evolution. The host rock conditions discussed in the **Data report** are groundwater composition, in-situ temperature, and in-situ stress. However, it is argued that the effective diffusivity and, especially, the porosity are relatively insensitive to host rock conditions.

Concerning the groundwater composition, which affects the porewater composition, the salinity may affect the degree of anion exclusion in the porous system. This is judged to have an insignificant effect on the porosity but a minor effect on the effective diffusivity. In the **Data report**, this is treated as data uncertainty, and the data provided should encompass not only reasonable groundwater

The time for gas breakthrough is determined by the failure time of the copper shell and the corrosion rate of the canister insert (see Section 13.5.2). At the time of breakthrough, half of the inventory of C-14 and Rn-222 is taken to be released immediately to the biosphere. The remaining gaseous inventory (and the Rn-222 that is produced) is then taken to be released together with the gas that is produced continuously. However, this release is neglected, since it will be insignificant in comparison with the pulse release. If the release occurs in the first 10,000 years (unlikely) the release of C-14 would be about 10 GBq. A release of Rn-222 would be about 25 GBq if the release occurred after 100,000 years.

In /SKB 2006g, a/ the calculated exposures from pulse releases of C-14 and Rn-222 are presented. C-14 may be released as methane (CH<sub>4</sub>) or carbon dioxide (CO<sub>2</sub>). It is assumed that if C-14 is released as methane from the repository, it will be oxidised to carbon dioxide by soil organisms. There are several alternatives regarding the fate of methane in its travel towards the surface:

1. Nothing happens and it will be released as gas to the atmosphere.
2. Microorganisms incorporate methane in the biosynthesis of other organic substances (i.e. not an oxidation). Eventually this organic carbon will be released by respiration to CO<sub>2</sub>. This may occur in the rock as well as in the regolith.
3. Other microorganisms utilise the carbon source and produce CO<sub>2</sub> (i.e. an oxidation). This may occur in the rock as well as in the regolith.
4. The produced CO<sub>2</sub> will either degass to the atmosphere or be used in the photosynthesis by aquatic or terrestrial algae/plants. This may occur in surface ecosystems.

Thus, if a pessimistic approach is taken, the release as CO<sub>2</sub> will at least be handled in the biosphere model before it is released to the atmosphere. Methane released to the atmosphere is unlikely to be utilised. However, the potentially increased mobility of a gas in the geosphere is not considered.

Radon is a noble gas and will not undergo chemical transformations. Two exposure cases are considered, one outdoors where radionuclides can be inhaled or consumed via uptake in plants in an area of 10,000 m<sup>2</sup>, subject to a wind speed of 2 m/s and a mixing height of 20 m, the other inhalation of radionuclides indoors in a house with a volume of 1,000 m<sup>3</sup> and a ventilation rate of 2 h<sup>-1</sup>. For C-14, exposure may occur via inhalation or ingestion, for Rn-222 only inhalation of Rn-222 and its radioactive daughter products needs to be taken into account. A summary of the results is given in Table 13-11. It is noted that no account for decay in transit from the repository to the surface is taken, making the results for Rn-222, with a half-life of only 3.8 days, further pessimistic.

If the gas pressure is built up during a period of glaciation, the hydrostatic pressure from the ice has to be added to the gas breakthrough pressure. This may lead to internal pressures of about 50 MPa inside the canister. If the retreat of the ice is rapid, this could lead to pressure drops of around 40 MPa and consequently 80% of the gaseous inventory would be instantaneously released.

The highest dose from a gas pulse of Rn-222 occurs in buildings. It is below the regulatory limits for an annual average life time risk for a repository, and it is considerably lower than the consequences of today's limit of 200 Bq/m<sup>3</sup> for radon in buildings in Sweden, which gives about 2 mSv/y.

**Table 13-11. Calculated annual mean life time risk from pulse releases of C-14 and Rn-222 for a single canister /SKB 2006g, a/.**

Pathway	C-14 (µSv) (10 GBq release)	Rn-222 (µSv) (25 GBq release)
Ingestion	0.036	–
Inhalation outdoors	4.4 · 10 <sup>-5</sup>	0.22
Inhalation indoors	0.0028	7.2

## **The time required to fill the canister with water**

In the **Data report** it is shown that for a hypothetical, circular penetrating defect with a radius of 2 mm it will take more than 20,000 years to fill the canister with water. To cover any shape of the defect, the delay time is pessimistically estimated to be at least 1,000 years, used as the pessimistic delay time in the hypothetical pinhole case analysed in Section 13.7.2.

The derivations of all these delay times neglects the effect of gas generated by corrosion of the insert by the penetrating water, which could lead to substantially longer delay times /Bond et al. 1997/.

## **The buffer's contribution to delaying water saturation and limiting the rate at which the canister is filled with water**

A prerequisite for water to enter into the canister is a fully water saturated buffer. The saturation times for both backfill and buffer are likely to range from a few tens of years to several thousand years, as a consequence of the rock properties (matrix hydraulic conductivity and presence and characteristics of fractures) at Forsmark. Consequently, in many deposition hole positions, no water would be expected to come into contact with the waste during the first 1,000 years even if there were an initial defect in the canister.

## **The containment function of the Zircaloy cladding**

According to the **Fuel and Canister process report**, available data suggest a life of the cladding tubes of at least 100,000 years. Although Zircaloy is highly resistant to uniform corrosion, due to its potential susceptibility to local corrosion in groundwaters and to hydrogen induced cracking, cladding is not assumed to constitute a barrier to radionuclide release from the fuel in SR-Site. However, even a cladding with small cracks or corrosion defects would offer a large mass-transport resistance for water to get into contact with the fuel and for dissolved radionuclides to exit into the canister void.

## **Radionuclide retention in the buffer**

The retention properties of the buffer are exemplified in the calculation cases with assumed complete losses of barrier functions in Section 13.7.3. A comparison between Figures 13-63 and 13-61 shows that the buffer has a considerable impact on near-field releases of many sorbing nuclides.

## **The host rock's favourable hydrogeological properties**

In addition to contributing to long saturation times mentioned above, the limited flow rates at the deposition positions limits the release rate of radionuclides from the near field if releases occur from the canister.

## **Radionuclide retention in the rock**

The retention properties of the rock are exemplified in the calculation cases with assumed complete losses of barrier functions in Section 13.7.3. A comparison between Figures 13-62 and 13-61 shows that the rock has a considerable impact on far-field releases of many sorbing nuclides.

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