



Working Report 2008-47

# Mass Balances and Energy Flows Reference Concept (Spent Fuel — Copper-Iron — Bentonite — Granite)

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December 2008

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## **Mass Balances and Energy Flows, Reference Concept (Spent Fuel–Copper-Iron–Bentonite–Granite)**

### **ABSTRACT**

In this work, a semi-quantitative analysis of mass and energy flows and balances in a deep repository of the KBS-3V type subject to a glacial cycle has been carried out. The energy flows and temperatures show the maximum temperature at the canister surface not to exceed the design temperature of 100 °C. If the measures taken to limit the water flow into the underground facilities are appropriate, the lifetime of the calcite buffer in the hydraulically conductive fracture zones was calculated to extend well beyond the operational phase of the repository. The results from hydrogeochemical model calculations in the backfill imply a long-term exchange of sodium for calcium in the clay component, if MX-80 bentonite is used. As this constitutes a potential threat to the swelling pressure of backfill in saline water environments, the physicochemical properties of a backfill should be carefully adjusted to meet its preplanned function. Despite short-lived episodes of oxygen-rich glacial water intrusion, the corrosion of the copper canister will likely be minor in the long term.

## **Massa – ja energiavirrat**

**Referenssikonsepti: käytetty polttoaine – kupari – rauta – bentoniitti – graniitti**

### **TIIVISTELMÄ**

Tässä työssä on tehty semikvantitatiivinen analyysi massa- ja energiavirroista ja tasapainoista KBS-3V tyyppisessä syvässä loppusijoitustilassa, joka on alttiina jääkausisyklille. Energiavirrat ja lämpötilat osoittavat, ettei maksimilämpötila kanisterin pinnalla ylitä 100 °C:n suunnittelulämpötilaa. Veden virtausta maanalaisiin tiloihin rajoittavien toimenpiteiden ollessa asianmukaiset, laskettiin kalsiittipuskurin elinajan hydraulisesti johtavassa rakovyöhykkeessä ulottuvan hyvin toimintavaiheen yli. Hydrogeokemialliset mallilaskut täyteaineessa viittaavat pitkäaikaiseen natriumin vaihtumiseen kalsiumiksi savikomponentissa, jos käytetään MX-80 bentoniittia. Koska tämä muodostaa potentiaalisen uhan täyteaineen paisuntapaineelle suolaisessa vesiympäristössä, täyteaineen fysikokemialliset ominaisuudet pitäisi säätää huolellisesti vastaamaan sille edeltä suunniteltuja toimintoja. Happirikkaan jääkausiveden lyhytaikaisista tunkeutumiskausista huolimatta kuparikapselin korrosio tulee olemaan vähäistä pitkällä tähtäimellä.

## **PREFACE**

This study is part of the NF-PRO project within the Sixth Framework Programme of the European Commission (Integrated Project NF-PRO, Contract No. FI6W-CT-2003-02389). This subproject has been co-funded by the European Commission and Posiva Oy.

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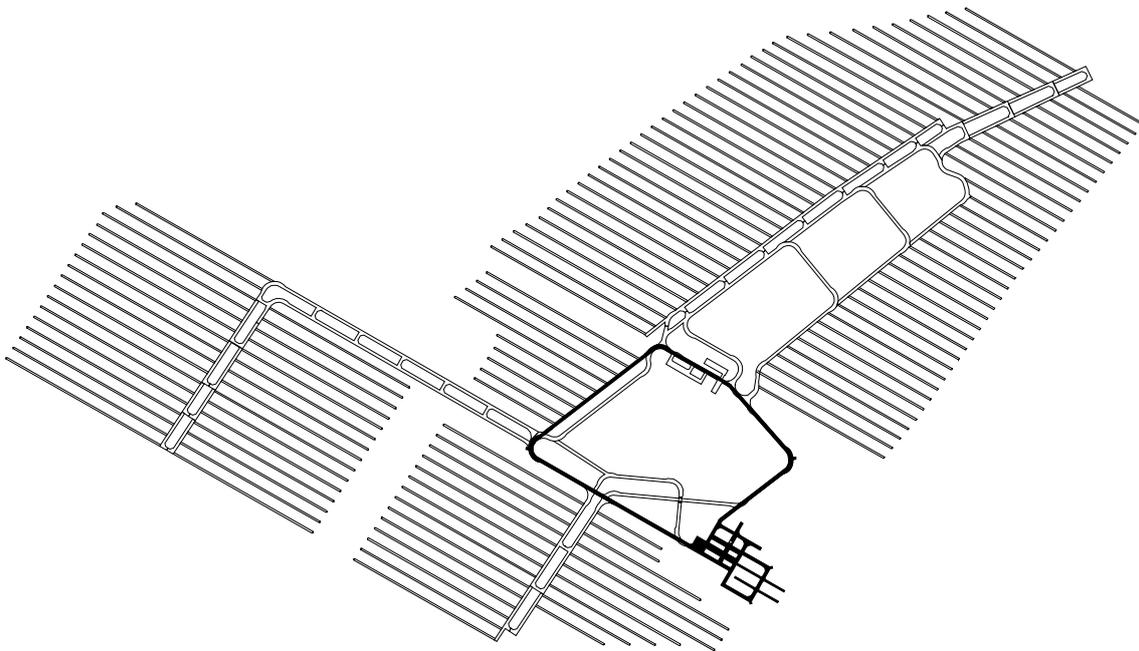
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## 1 GENERAL

In this work, which is part of the EU's integrated NF-PRO project, a semi-quantitative analysis of mass and energy flows and balances in a deep repository of the KBS-3V type has been carried out. Complementary model calculations for the long-term geochemical evolution of a deposition tunnel subject to an assumed glacial cycle have been carried out by Luukkonen and Pitkänen (2006).

### 1.1 Deep repository

The layout of the repository at the depth of 400 m to 700 m in crystalline bedrock has two alternatives: the one- or the two-storey construct. The general layout of a KBS-3V one-storey repository is shown in Figure 1-1.



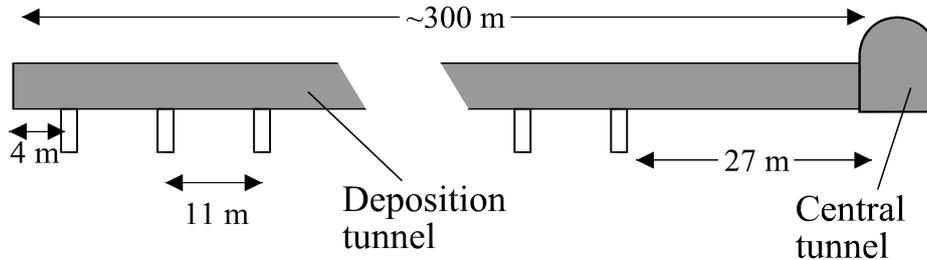
*Figure 1-1. General layout of a one-storey repository.*

The actual disposal of spent-nuclear-fuel canisters is a continuous process. During the operational phase, the disposal tunnels are excavated from the central tunnel. The length of a disposal tunnel will be about 300 m. The number of canisters containing the Olkiluoto BWR fuel will be about 24 and the Loviisa PWR fuel about 31 per tunnel. This is due to the different heat production rates of the respective fuels.

The distance between the nearest canister in the disposal tunnel and the central tunnel will be about 27 m. Consequently, mass transfer even to the nearest canister from the central tunnel will take a very long time. The separation distance between canisters in a

deposition tunnel will be about 11 m for the Olkiluoto fuel and the spacing between parallel deposition tunnels about 25 m.

The layout of a disposal tunnel is shown in Figure 1-2 and the properties of the tunnels in Table 1-1.



**Figure 1-2.** *Emplacement of canisters in a deposition tunnel for the Olkiluoto BWR fuel.*

**Table 1-1.** *Properties of deposition tunnels.*

Section	Width (m)	Height (m)	Area (m <sup>2</sup> )	Total length (m)
Central tunnel	6.00	6.0	32.7	7 500
Deposition tunnel	3.50	4.40	14.00	41 000

## 1.2 Deposition hole buffer material

The deposition holes will be filled with a buffer of bentonite clay. There will be a 10-mm air gap between the lateral sides of the canister and buffer, and a 35-mm water gap between the wall of the hole and the buffer. Without the gaps, the volume of the bentonite is 13 m<sup>3</sup> for deposition holes with canisters for the Olkiluoto BWR fuel, and 10 m<sup>3</sup> for deposition holes with canisters for the Loviisa PWR fuel. The dry density of bentonite is assumed to be 1 700 kg/m<sup>3</sup>.

## 1.3 Backfilling concept

The general concept for the backfilling of the repository is by way of a mixture of crushed rock and bentonite. In this study and, the following parameterization is used in the calculations:

- the buffer in the deposition holes consists 100% of bentonite,
- deposition tunnels are backfilled with a mixture consisting of 30 wt% bentonite and 70 wt% crushed rock, and
- the rest of the repository is backfilled with a mixture consisting of 15 wt% bentonite and 85 wt% crushed rock.

The dry density of the backfill is assumed to be  $1\,700\text{ kg/m}^3$ , as in Luukkonen and Pitkänen (2006).

## 2 OPERATIONAL PHASE

During the operational phase, about 40 canisters will be emplaced within 10 months. Consequently, one deposition tunnel for the Olkiluoto BWR fuel will be used for about 6 months (24 canisters), and, therefore, a typical time for the emplacement of one canister is one week.

The engineering and stray materials have been identified and their remaining quantities after backfilling have been estimated for different design alternatives and construction solutions (Hjerpe, 2004). The results show that the total remaining quantity will be in the range from 13 000 to 28 000 tonnes. Cement is the dominant component, with about 75% to 85% of the total. Steel, organic material, aluminium, and zinc are other important materials.

The unit volume in which the mass and energy will be analyzed in detail is about 170 m<sup>3</sup>, equivalent to the sum of the volume of an 11-m length of tunnel (154 m<sup>3</sup>) and the corresponding deposition hole (19 m<sup>3</sup>). The saturation degree of bentonite and backfill is assumed to be 58% after emplacement. The most important natural species trapped in the repository cell of 170 m<sup>3</sup> is oxygen. The porosity of bentonite is 43% and backfill 36%. Thus, the amount of trapped air is about 2.35 m<sup>3</sup> in the buffer and 23.5 m<sup>3</sup> in the backfill and 1.60 m<sup>3</sup> in the air gaps (about 27 m<sup>3</sup> in total). Thus, the total amount oxygen in this volume is about 250 moles. This amount of oxygen may corrode 500 mol (32 kg) of copper to form tenorite (CuO), or 1 000 mol (64 kg), if cuprite (Cu<sub>2</sub>O) is formed (provided no other oxygen-consuming reactions, such as oxidation of pyrite in the buffer, take place). The total amount of copper in a deposition hole will be 7 500 kg, and, therefore, the effect of trapped oxygen on the general corrosion of copper will be of minor importance. However, upon deglaciation, the oxygen dissolved in meltwater may cause a significant flow of oxidative agents towards the repository, which could have implications for the canister corrosion (see Chapter 5).

After the emplacement of a canister, the unit cell contains engineering materials as shown in Table 2-1.

*Table 2-1. Materials in a unit cell after the emplacement of an Olkiluoto canister.*

<b>Material</b>	<b>Mass (kg)</b>	<b>Volume (m<sup>3</sup>)</b>
Backfill	260 000	154
Bentonite (gaps excluded)	22 000	13.0
Olkiluoto canister	24 500	4.2
Air gaps	2	1.6
Air total	35	27
Oxygen	8 (250 mol)	5.7 (NTP)
Water (58% saturation)	35 000	35
Low-alkali cement at the bottom of the hole	46	~0.02
Other cement in the deposition tunnel	~0	~0 (removed)

### 3 PRE- AND POST-CLOSURE TRANSIENT PHASE ON A SITE SCALE

When a deposition tunnel has been backfilled, the groundwater will enter the tunnel. It is possible that some water enters the tunnel during the emplacement period of a few months, but this amount will be small compared to the pre-closure-phase inflow.

The large-scale analysis in this chapter has been made for the Olkiluoto site using the FEFTRA software (Löfman and Mészáros, 2002), which is a thermo-hydraulic (TH) model, capable of accounting for thermal effects and upconing of deep saline water.

The model assumptions were:

- the repository will be open for 100 years,
- 90% of the repository will be closed, meaning that the canisters are emplaced and tunnels are backfilled,
- 10% of the repository will be open for 100 years and closed thereafter (see Figure 3-1), and
- the porosity of rock and the repository is set at 0.01%.

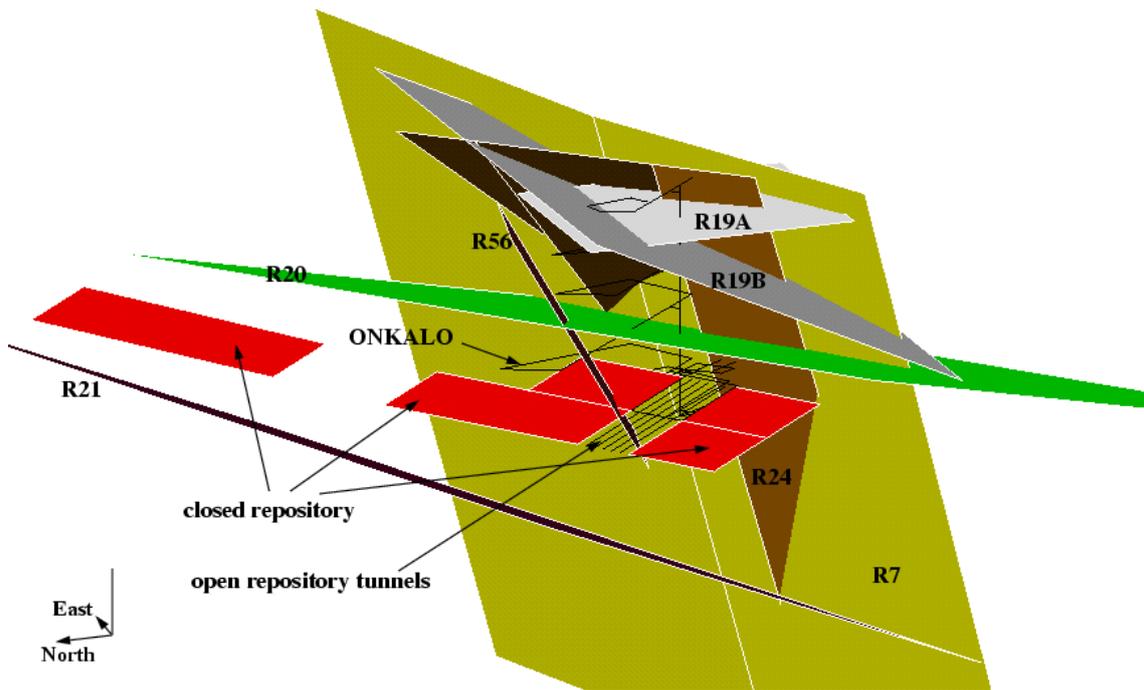
The density of the flowing groundwater is affected by the temperature and salinity. Therefore, hydraulic gradients due to variations in water density will be established. Also, the viscosity of water decreases with temperature, which will increase the mobility of the groundwater. These phenomena were taken into consideration in this study.

Thermal effects were not taken into account in Vieno et al. (2003) and, further, only the ONKALO research tunnel was assumed to exist, as the work focussed on the disturbances caused by the construction and operation of ONKALO. The amount of disposed fuel and the corresponding production of decay heat as a function of time are depicted in Figure 3-2.

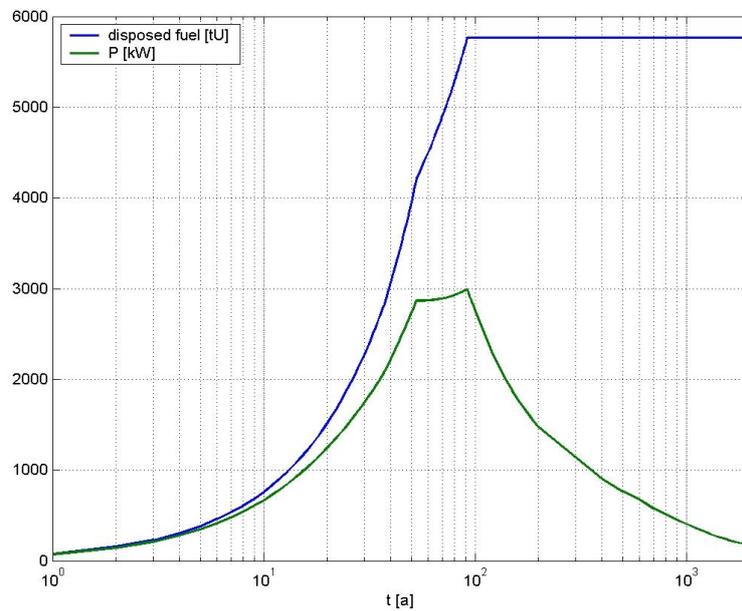
The results shown in Figures 3-3 to 3-6 for the one-storey layout may be interpreted as follows:

- the maximum temperature rise at the repository level will be about 50 °C after about 100 years, and
- the maximum salinity will be about 40 g/l at the repository level.

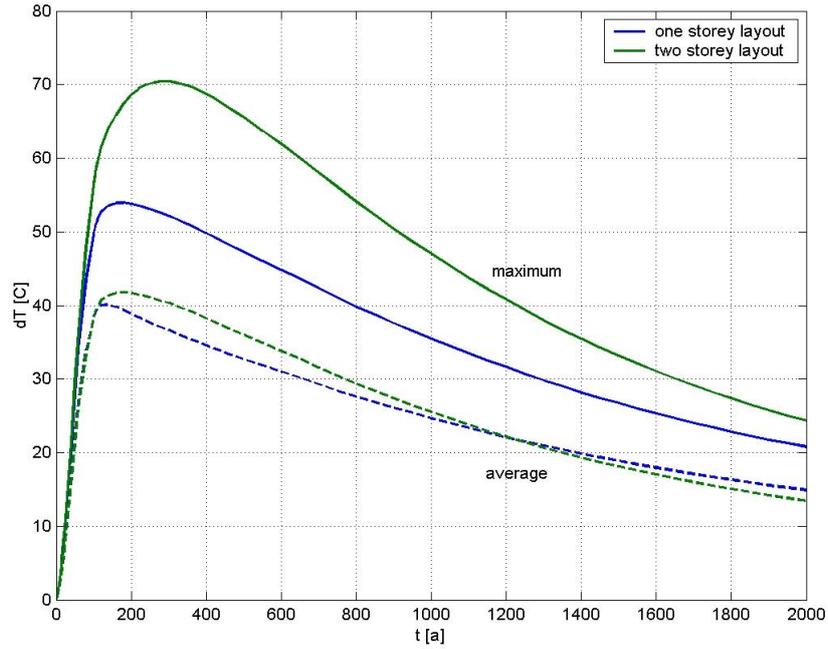
After the closure of ONKALO, the salinity distribution tends towards its natural undisturbed state (Figure 3-4). After 200 years, the disturbances have more or less been evened out.



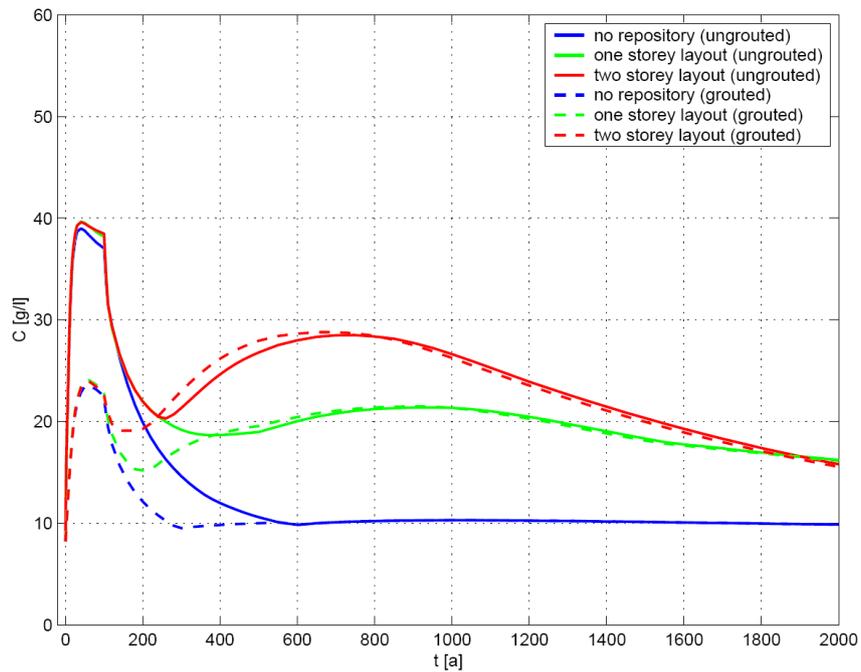
*Figure 3-1. Model geometry of the repository and fracture zones (R7, etc.)*



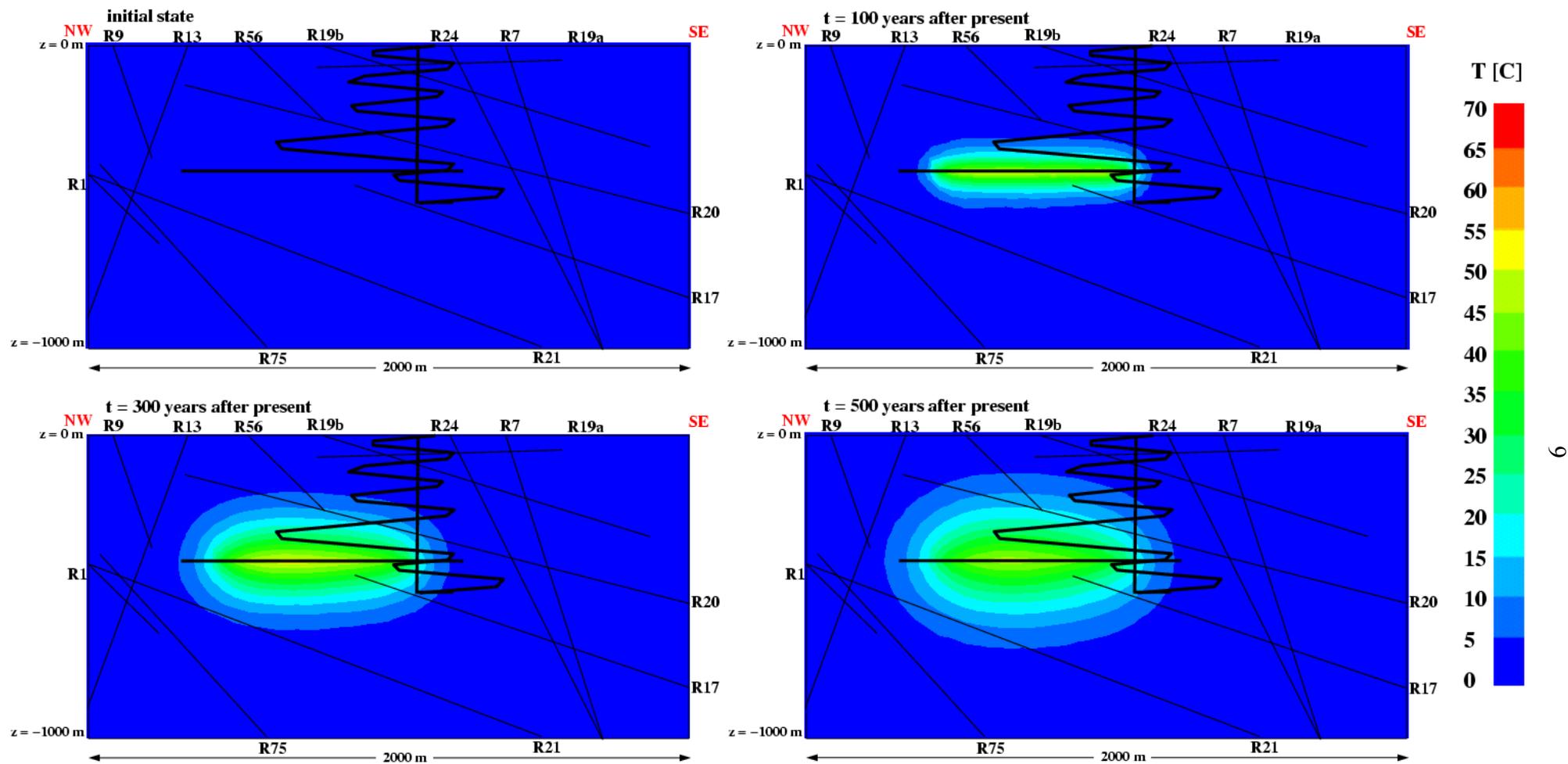
*Figure 3-2. Disposed fuel and production of decay heat as a function of time in the repository.*



**Figure 3-3.** Temperature rise in the repository (blue line: the one-storey layout).



**Figure 3-4.** Predicted evolution of maximum groundwater salinity [g/l Total Dissolved Solids (TDS)] at a repository depth of 420 m for the one- and two-storey layouts (from Löffman, 2005). The effect of tunnel grouting shown as broken lines.



*Figure 3-5. Temperature rise at vertical cross section; at initial state, 100 years (instantaneous closure of repository), 300 years and 500 years.*

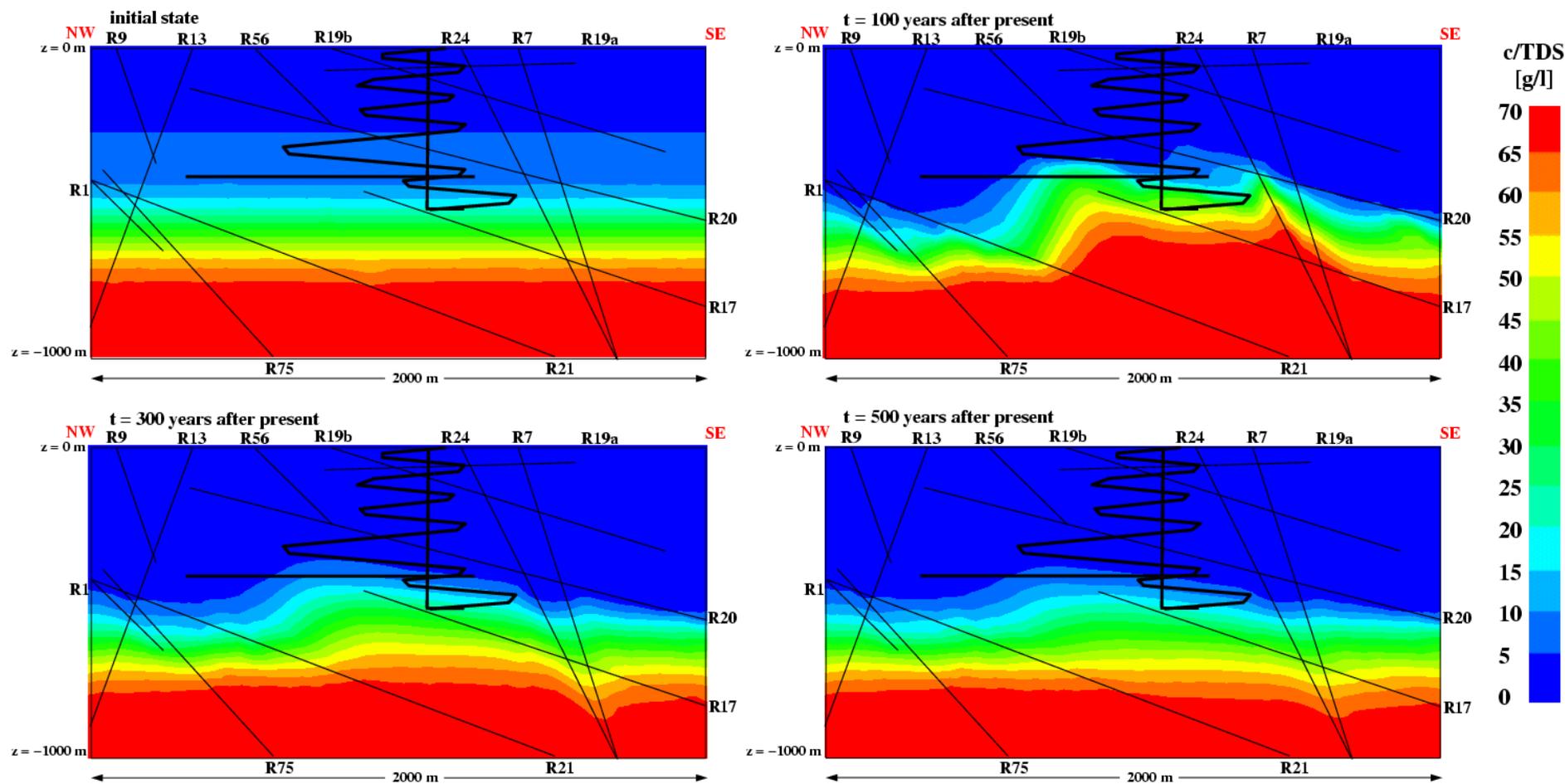


Figure 3-6. Salinity at vertical cross section; at initial state, 100 years (instantaneous closure of repository), 300 years and 500 years.

## 4 UNIT CELL ANALYSIS SHORTLY AFTER REPOSITORY CLOSURE

The balances and flows are grouped as follows:

- the flows and balances inside the 170-m<sup>3</sup> unit cell correspond to a single deposition hole,
- the external mass flows, and
- man-made stray and engineering materials that are treated separately from natural species (oxygen and salt).

The species of interest are:

- oxygen and acidic species,
- total dissolved solids (TDS), i.e., salinity,
- alkalinity originating from cement, and
- water.

The mass transfer from water flowing in a fracture intersecting a deposition hole can be estimated by applying a boundary layer (film) resistance model (Vieno and Nordman, 1999),

$$Q_{bl} = 2\sqrt{2DQ_f 2b_v}, \quad (4-1)$$

where

- $Q_{bl}$  is the effective flow rate (m<sup>3</sup>/s),
- $D$  is the diffusion coefficient in water (m<sup>2</sup>/s),
- $Q_f$  is the flow rate of groundwater in the fracture (m<sup>3</sup>/s),
- $2b_v$  is the volume aperture of the fracture (m).

Equation 4-1 has been used to derive the mass transport coefficients of TILA-99 from a deposition hole to a fracture in the range from 0.2 to 5 l/a. The total effective flow rates through unit cell (mostly via deposition tunnel) are from 100 to 2 000 l/a.

### 4.1 Oxygen and host rock fracture fillings

Infiltration of low-pH meteoric water containing oxygen into the disposal level and its effects on calcite and pyrite fracture coatings have been analysed by Luukkonen et al. (2004). These calculations give a rough idea of the type of water that will be infiltrating into a deposition hole. The coupling of flow and reactions with the soil water was studied with flow velocities of 0.1, 1.0 and 10 l/h, and with the rain water with velocities of 1.0 and 10 l/h.

The calculations were based on calcite and pyrite dissolution kinetics and transport of dissolved oxygen, carbon dioxide and bicarbonate, but, otherwise, the exercise was greatly simplified. The variation of flow rate was found to have an impact on the calcite buffer consumption rates. Increasing the flow by one order of magnitude caused particularly large erosion in the calcite buffer, because even with a flow rate of 10 l/h, calcite soon equilibrated with the flowing water. According to the model results, every

attempt to limit the water flow into the underground facilities has a great effect especially on the lifetime of the calcite buffer in the hydraulically conductive fracture zones. Considering the remarkable variation in pH, which depends on the availability of calcite, it must be pointed out that the exercise focussed on a simplified system. Factors other than mere availability of calcite buffers are also likely to affect the pH levels. As an example, weathering of silicates, especially in the top soil layer, and microbial processes in general will likely have significance to the pH and redox levels of the down-flowing water. Because the simulations were run only for 50 years onward, they do not give an exact idea about the amount or distribution of buffers at the time of repository closure. However, the current exercise indicated that these kinds of calculations, but with more realistic boundary conditions, can be of use.

The oxygen concentration of infiltrating water at the ground level was 10 mg/l (Luukkonen et al., 2004). According to the simulations at the repository depth, the oxygen concentration will be about 2 mg/l after 30 years of infiltration of soil water with a flow rate of 1 l/h. Thus, the amount of oxygen in infiltrated water during the saturation phase (max. 54 g in 27 m<sup>3</sup> of water) is small in comparison to the amount of residual oxygen (8 kg).

## 4.2 Thermal energy and resaturation

The main result from the preliminary analysis carried out by Lempinen (2005) was that the resaturation of a KBS-3V deposition hole will take about 30 years, which is somewhat longer than the results from previous simulations found in the open literature. In fact, based on model calculations, the uncertainty in the duration of the resaturation period seems to be particularly great.

In addition to analyses by Löfman (2005) (see Chapter 3), the thermal evolution of the repository has been calculated by Ikonen (2003) and Lempinen (2005). In both of these studies, the initial thermal power of a canister was 1.7 kW, which represents a BWR canister with a precooling time of 38 years for the fuel. The decay heat in the canisters is halved every 30 years and, thus, the maximum temperature in the fuel, canister and the near-field is reached in a few decades. However, the increased temperature in the host rock around the repository will persist for a few thousand years (see e.g. Figure 3-3).

The reference case of Ikonen (2003) was the following:

- the thermal conductivity of bentonite was set at 1.0 W/m/K, which is close to the value of fully saturated bentonite of 1.3 W/m/K,
- the saturation process itself was not modelled,
- for the host rock, a thermal conductivity of 2.62 W/m/K was adopted,
- the maximum temperature in a central tunnel will be about 40 °C after about 300 years, and
- the maximum temperature reached at the canister surface will be 90 °C after about 50 years.

The assumptions and preliminary results from the finite-element-based thermo-hydro-mechanical (THM) model of Lempinen (2005) were:

- The host rock temperature at some distance from the buffer-rock interface was fixed at 45 °C.
- The effects of moisture (or vapour) transport, swelling pressure and saturation degree of bentonite were taken into account, contrary to Ikonen (2004)
- The maximum temperature reached at the canister surface will be between 74 °C and 84 °C.

The final results will be published within the WP 3.3 of the NF-PRO project (Lempinen, 2005).

The three models used in Finland are different from each other and, therefore, the problem lies in how to interpret and compare the obtained results. Here, we choose to look at the results of Ikonen (2004) in more detail. During the first 50 years, the average heat power of a BWR canister will be about 1.4 kW. Consequently, about 2.2 TJ of energy will be produced. In a unit cell containing one canister, the surface temperature of the canister will be 90 °C, the ambient temperature 10 °C, the volume of a canister 4.2 m<sup>3</sup>, and the heat capacity 2.4 MJ/m<sup>3</sup>/K. In the bentonite with a heat capacity of 2.2 MJ/m<sup>3</sup>/K, the temperature rise will be less than 60 °C. In the backfill, with a heat capacity of 1.75 MJ/m<sup>3</sup>/K, the temperature rise will be less than 50 °C. Thus, the total energy contained in the backfill and the bentonite buffer will be at most 15 GJ.

To conclude, most of the thermal energy produced will dissipate from a canister or tunnel vertically within a distance of at least 100 m. The energy produced in 50 years will be spread over a rock volume of at least 30 000 m<sup>3</sup>. The equivalent increase in rock temperature will be about 40 °C for the assumed heat capacity of rock of 2.15 MJ/m<sup>3</sup>/K, which can also be seen from Figure 3-3 or Figure 40 in Ikonen (2003). The increase in temperature is consistent with the produced energy of 2.2 TJ.

### 4.3 Alkaline plume

The potential effects of a high-pH plume originating from cementitious grouting are discussed in Vieno et al. (2003).

The total amount of cement remaining in ONKALO at the time of backfilling was estimated to be at most 5 400 tonnes. Approximately 3 000 tonnes would originate from rock grouting and the rest mainly from shotcrete. Most of the cement would remain in the access tunnel and shaft as they constitute the main part of the excavations for ONKALO. The 5 400 tonnes of cement could release about  $3 \cdot 10^7$  mol of hydroxide ions on total chemical exhaustion.

In general, it is fairly unlikely to encounter a plume with pH significantly higher than 11 at a distance of several tens of metres from the nearest grouted location. This corresponds to the minimum separation distance of ONKALO excavations and disposal tunnel. In addition, mass transfer from a fracture intersecting a deposition hole is limited

by diffusion and boundary layer (film) resistance (Eqn. 4-1). Also, the buffering capacity of bentonite for hydroxide ions is at least 0.5 mol/kg of clay. A repository of the KBS-3V type for 6 000 tonnes of spent fuel contains about 60 000 tonnes of bentonite clay in the buffer in the deposition holes. The tunnel backfill may contain similar or greater amounts of bentonite or other swelling clays. In theory, the cement remaining in ONKALO could alter all the bentonite in the deposition holes if transported into the deposition holes, evenly distributed among them and if there were no other buffering reactions operative. In other words, the clay in the buffer alone could neutralize all the hydroxide released from the cement remaining in ONKALO. However, it is not possible that all the cement in ONKALO, for example within and above the major sub-horizontal fracture zones R19 and R20 (see Figure 3-1), could be transported without any buffering and/or dilution into the canister deposition holes. Consequently, the probability that an alkaline plume could intrude a deposition unit cell is small, but further research on associated uncertainties should be carried out.

#### 4.4 Salinity (TDS)

After the closure of a deposition hole or tunnel, the saline Olkiluoto groundwater begins to resaturate a deposition hole. The maximum estimated salinity will be about 40 g/l. Thus, assuming a saturation degree of 58%, 18% of bentonite and 15% of backfill will be resaturated with saline water. In addition, the 35-mm gap between bentonite and rock will be filled with saline water. According to Lempinen (2005), the resaturation time will be some 30 years, and, therefore, the water infiltrating to at least one deposition hole will have an average salinity of less than 40 g/l. As in Luukkonen and Pitkänen (2006), the concentration is assumed to be 20 g/l.

#### 4.5 Material balance after resaturation

In Table 4-1, the quantities of different masses in a unit cell after the saturation phase of about 30 years are estimated. The estimates are very qualitatively based on the discussion in this chapter.

*Table 4-1. Materials in a unit cell after the saturation phase, i.e. about 30 years after canister emplacement.*

Material	Mass (kg)	Volume (m <sup>3</sup> )
Backfill	260 000	154
Resaturated bentonite	22 000	14.6
Olkiluoto canister	24 500	4.2
Oxygen	8 (250 mol)	Dissolved in pore fluid
Water	63 000	63
TDS from infiltrated water (~27 m <sup>3</sup> )	560	n.a.
Low-alkali cement at the bottom of the hole	46	~0.02

## 5 LONG-TERM GEOCHEMICAL MASS BALANCES

A simple analysis of long-term mass balances is carried out on the basis of model calculations by Luukkonen and Pitkänen (2006) and on the climate scenarios analyzed in Cedercreutz (2005).

The aim of the climate-scenario study of Cedercreutz (2005) was to search in the open literature for the latest research concerning future climate scenarios in order to present relevant climate evolution for Olkiluoto as far as possible into the future – in practice, up to 125–225 ka AP. Emphasis was put on examining permafrost occurrence and shoreline displacement during a glacial cycle. Predictions of a future climate inherently include uncertainties. From the available scenarios, one was chosen which assumes that the burning of fossil fuel will contribute to a small increase in the natural CO<sub>2</sub> concentration in the atmosphere. This anthropogenic contribution will add 850 ppmv (parts per million by volume) to the natural concentration at 325 years after present (AP), for a total of 1 146 ppmv. According to this scenario, glaciation will not affect Olkiluoto until at ~200 ka AP, at which time there will still be an anthropogenic contribution of 50 ppmv of CO<sub>2</sub> left.

Table 5-1 presents the assumptions made in the present analysis.

*Table 5-1. Assumed flow rate through unit shell and salinity of inflowing water during different time periods.*

Time period (ka)	Description	Flow rate (l/a)	Salinity (g/l) <sup>†</sup>
0–10	Present island	100	20
10–200	Mainland	100	5.8
200–202	Advancing ice sheet	1 500	11.4
202–225	Under ice	300	4.4

<sup>†</sup> From Luukkonen and Pitkänen (2006)

In Table 2-2 of Luukkonen and Pitkänen (2006) are presented the amounts of material in a unit volume defined in a hydrogeochemical model (11 kg of backfill) at the beginning of calculation.

### 5.1 From resaturation to 10 ka AP

During this phase, it is assumed that saline groundwater (TDS 20 g/l) flows through the unit cell at a rate of 100 l/a. The result is that after some time, salt (TDS) and oxygen will be removed from the unit cell. With an effective outflow rate from the unit cell of approximately 100 l/a, it can be estimated that, of the dissolved salt and oxygen in the water, a fraction of about 0.16% ( $= 0.1 \text{ m}^3/63 \text{ m}^3$ ) will be removed annually. Hence, within 2 000 years, the concentration decreases to about 4% of the original, and within 10 000 years to zero, even without chemical reactions. The material balance presented in Table 5-2 for a unit cell at 10 ka AP is assumed to persist thereafter.

*Table 5-2. Materials in a unit cell after 10 ka.*

<b>Material</b>	<b>Mass (kg)</b>	<b>Volume (m<sup>3</sup>)</b>
Backfill	260 000	154
Resaturated bentonite	22 000	14.6
Olkiluoto canister	24 500	4.2
Oxygen	0	0
Water	63 000	63
Low-alkali cement at the bottom of the hole	46	~0.02

Some of the results from the model calculations presented in Table 6-1 of Luukkonen and Pitkänen (2006)<sup>1</sup> for this period of time are:

- About 50% of sodium in the backfill will be replaced by calcium from the groundwater. The molar fractions of exchangeable sodium and calcium are predicted to be approximately equal (49% and 50%, respectively), the initial molar fraction for exchangeable sodium being 91%. Magnesium and potassium play the role of minor exchangeable cations; their mole fractions never exceeded 5% and 1%, respectively, in any of the calculations.
- The accumulated amount of sulphide, the only corrosive agent identified in deep groundwaters to initiate and sustain corrosion of copper, is 22.8 kg. It results from the groundwater input and consumption of organic matter by sulphate reduction, and from subsequent precipitation within a unit volume. In relation to the initial pyrite (FeS<sub>2</sub>) impurity of 0.3% by weight<sup>2</sup> in MX-80 bentonite (Müller-Vonmoos and Kahr, 1983), this amount probably does not add to the corrosion penetration of a canister owing to the low solubility of sulphide in anoxic reducing conditions and strong anion exclusion in compacted smectite clay. After this stage, no net mass transfer of sulphide is predicted to take place [Table 5-2, and Tables 6-2 to 6-4 in Luukkonen and Pitkänen (2006)].

The cation exchange to calcium form constitutes a potential threat to the swelling pressure of backfill in saline water environments. This is due to the fact that the density of the backfill from the outset may not be high enough to sustain hydration swelling of the material. Instead, the swelling is governed by the interaction of electric double layers, the magnitude of which is sensitive to changes in the groundwater salinity. High salinities render this interaction non-existent, which results in the loss of swelling pressure. However, this problem is avoided in the highly compacted bentonite buffer, provided its density remains high enough despite possible minor erosion or dilution into the adjacent host rock, because the swelling is almost exclusively dictated by the hydration of smectite surfaces and exchangeable cations. Consequently, the swelling is insensitive to groundwater salinity, and the anticipated partial exchange of sodium for calcium could yield an increased swelling pressure.

<sup>1</sup> The calculations did not consider the effects of e.g. cementitious material.

<sup>2</sup> This amounts to 66 kg of pyrite in a compacted MX-80 buffer in a single deposition hole.

## 5.2 Mainland stage, 10–200 ka AP

During this phase, it is assumed that brackish groundwater (TDS 5.8 g/l) will flow through the unit cell at a rate of 100 l/a. The results of model calculations are shown in Table 6-2 of Luukkonen and Pitkänen (2006) and summarized below:

- A slight decrease and increase in the molar fraction of sodium and calcium, respectively, in the exchanger phase is predicted (41% and 55%, respectively).
- The system becomes somewhat more reducing. The fate of dissolved sulphur from the incoming groundwater is to precipitate as pyrite.

## 5.3 Advancing ice sheet, 200–202 ka AP

During this phase, it is assumed that brackish groundwater (TDS 11.6 g/l) will flow through the unit cell at a rate of 1 500 l/a. The results of model calculations are shown in Table 6-3 of Luukkonen and Pitkänen (2006) and summarized as follows:

- At the end of this epoch, the molar fractions of exchangeable sodium and calcium are 53% and 43%, respectively. Accumulation of sodium occurs, because the relatively high concentration of  $\text{Na}^+$  in the inflowing solution is preferred by the exchanger phase to  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , which are subsequently released into the pore solution.
- The system is characterized by small changes in its redox state.

## 5.4 Under ice, 202–225 ka AP

During this phase, the brackish groundwater (TDS 4.4 g/l) is assumed to flow through the unit cell at a rate of 1 500 l/a. The results of model calculations shown in Table 6-4 of Luukkonen and Pitkänen (2006) are:

- At the end of the simulation, the molar fractions of exchangeable sodium and calcium are 36% and 59%, respectively.
- Similarly to the previous stage, the redox state is subject to small variations.

## 5.5 Total, 0–225 ka AP

Table 5-3 lists the calculated geochemical net mass balance in the backfill over the whole time period of 225 ka. For a detailed discussion of the modelling procedure and results, see Luukkonen and Pitkänen (2006). It can be seen from Table 5-3 that the only species calculated to accumulate in the backfill are calcium and sulphide. The role of calcium is to replace exchangeable sodium in the exchanger phase, whereas all the soluble sulphide from the groundwater will precipitate as pyrite. The bulk of cation exchange will take place during the first 10 ka.

**Table 5-3.** Calculated mass balance in the backfill for the total calculatory time period, 0-225 ka AP. Positive and negative values indicate, respectively, increase and decrease in a unit volume.

Species	mol	kg
HCO <sub>3</sub>	-4 303	-262.6
SO <sub>4</sub>	-1 371	-131.7
HS	691	22.8
Fe <sub>tot</sub>	-3.7	-0.2
Cl	-6 721	-238.2
SiO <sub>2</sub>	-56	-3.4
Na	-31 287	-719.3
K	-908	-35.5
Ca	9 899	396.8
Mg	-140	-3.4

## 5.6 Beyond 225 ka AP

Prior to restoration of present-day temperate climatic conditions, voluminous discharges of meltwater upon deglaciation will occur. If this oxygen-rich water reaches the repository depth and infiltrates the backfill, it may increase the solubility of sulphidic precipitates. This may be counteracted by the increased anion exclusion in such diluted groundwater environments, however. Furthermore, the effects of such relatively short-lived episodes of meltwater intrusion will likely be of limited extent, since no clear evidence exists to date that oxidizing water has ever penetrated to great depths in regions, which have undergone repeated glaciations. Over time, the groundwater composition will revert to its original undisturbed condition. The backfill and buffer are likely to continue undergoing small fluctuations in their exchanger phase cationic compositions.

## 6 DISCUSSION AND CONCLUSIONS

In this work, a semi-quantitative analysis of mass and energy flows and balances in a deep repository of the KBS-3V type subject to a glacial cycle has been carried out. Complementary model calculations for the long-term geochemical evolution of a deposition tunnel are presented in a separate contribution (Luukkonen and Pitkänen, 2006).

The energy flows and temperatures were estimated based on results from three previous studies, which are fairly consistent, for example, in that they show the maximum temperature at the canister surface not to exceed the design temperature of 100 °C for the KBS-3V concept.

Infiltrating oxygen and carbon dioxide have the potential to consume the chemical calcite buffer in fracture fillings of the host rock, when the repository is open. If the measures taken to limit the water flow into the underground facilities are appropriate, the lifetime of the calcite buffer in the hydraulically conductive fracture zones was calculated to extend well beyond the operational phase of the repository (Luukkonen et al., 2004).

The results from simplified hydrogeochemical model calculations in the backfill imply a long-term exchange of sodium for calcium in the clay component, if MX-80 bentonite is used. As this constitutes a potential threat to the swelling pressure of backfill in saline water environments, the physicochemical properties of a backfill should be carefully adjusted to meet its preplanned function.

Despite short-lived episodes of oxygen-rich glacial water intrusion, the contribution of sulphide, calculated to precipitate in the backfill during the first 10 ka, to the corrosion of the copper canister will likely be minor in the long term.

Attainment of a comprehensive understanding, let alone quantification, of long-term mass and energy balances and flows is realistically beyond the realms of possibility for any project. The present work should be considered as a first attempt made to tackle this complex issue for the KBS-3V repository.

## REFERENCES

- BIOCLIM. 2001. Deliverable D3: Global climate features over the next million years and recommendation for specific situations to be considered.
- BIOCLIM. 2003. Deliverable D7: Continuous climate evolution scenarios over Western Europe (1000 km scale).
- Cedercreutz, J. 2004. Future climate scenarios for Olkiluoto with emphasis on permafrost. Report POSIVA 2004-06.
- Hjerpe, T. 2004. Engineering and stray materials in the repository for nuclear waste – Estimation of quantities after backfilling. Posiva Working Report 2003-72.
- Ikonen, K. 2003. Thermal analyses of spent nuclear fuel repository. Report POSIVA 2003-04.
- Lempinen, A. 2005. Thermo-Hydro-Mechanical analyses of a KBS-3V deposition hole. Deliverable 3.3.3 of RTDC 3.
- Luukkonen, A., Pitkänen, P., and Partamies, S. 2004. Significance and estimations of lifetime of natural fracture mineral buffers in the Olkiluoto bedrock. Posiva Working Report 2004-08.
- Luukkonen, A., and Pitkänen, P. 2006. Geochemical mass balances in a KBS-3V unit volume. To be published as a Posiva Working Report.
- Löfman, J., and Mészáros, F. 2002. Integration of equivalent continuum and discrete fracture network approaches in groundwater flow modelling – Preliminary study. Posiva Working Report 2002-39.
- Löfman, J. 2005. Simulation of hydraulic disturbances caused by the decay heat of the repository in Olkiluoto. Report POSIVA 2005-07.
- Müller-Vonmoos, M., and Kahr, G. 1983. Mineralogische untersuchungen von Wyoming bentonit MX-80 und Montigel. Nagra Report NTB 83-12.
- Vieno, T., and Nordman, H. 1999. Safety assessment of spent fuel disposal in Hästholmen, Kivetty, Olkiluoto and Romuvaara – TILA-99. Report POSIVA 99-07.
- Vieno, T., and Nordman, H. 2000. Updated compartment model for near-field transport in a KBS-3 type repository. Posiva Working Report 2000-41.
- Vieno, T., Lehtikainen, J., Löfman, J., Nordman, H., and Mészáros, F. 2003. Assessment of disturbances caused by construction and operation of ONKALO. Report POSIVA 2003-06.